Arsenic concentration and speciation in infant formulas and first foods.

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INTRODUCTION

Arsenic (As) is a ubiquitous metalloid, typically present at low concentrations in rocks, soils, and natural waters. The notable toxicity of As has led to its widespread anthropogenic use in insecticides and herbicide, first as an inorganic compound then later as the less toxic organic As compounds. Use of As in agriculture, industry, and As wastes from mining operations has led to localized areas of severe soil contamination which pose immediate human health risks from contaminated water, dust, or soil particles. More insidious, though, and affecting many millions more people, is exposure to "natural" geogenic As, resulting in both increased soluble As species in drinking water [1-3] and agricultural soils [4,5]. The detrimental health effects of As exposure via drinking water are well accepted and have led to a lowering of environmental regulatory limits to 0.01 mg [l.sup.-1] promoted by both the World Health Organization (WHO) and the U.S. Environmental Protection Agency (EPA) [6]. The exposure of millions of people to elevated levels of geogenic As in drinking water is happening now in the Asian subcontinent, particularly in Bangladesh but also in India, Vietnam, and Cambodia [7]. Indeed, anywhere where access to drinking water is via groundwater wells there is the potential for elevated As depending on the prevailing geology and subsurface biogeochemical conditions.

Although As is not generally readily taken up by crops or transported to the edible parts, a notable exception is rice, a staple food that can take up As from soil and transport it to the grain [4,8-10]. The magnitude of this uptake varies widely between cultivars, but the ability to take up elevated concentrations of As (in comparison with other cereal crops) appears to be a trait found in the entire rice germplasm. Elevated As concentrations in rice (relative to other food sources) were first reported in 1999 [11]. However, it is only recently that the potential human health implications for populations consuming rice-based diets has been fully appreciated, largely through the work of Meharg and colleagues in identifying food products containing As, quantifying and speciating As in these products, and showing that As intake through food can be equivalent to or greater than drinking water at the current safe drinking water limit [4,9,12-19].

Speciation of As in food products is necessary because of the differential toxicity of different As compounds. Organic As compounds, such as arsenobetaine found in seafood, are nontoxic and can be consumed without health concerns while others, such as inorganic As(III) and As(V) are toxic and pose a potential health risk [20]. Depending on growing conditions, rice contains predominantly dimethyl-arsenic acid (DMA) or inorganic As [14,21]. Inorganic As is more toxic than the methylated As species. While direct speciation of the solid compound would be preferable, current analytical techniques do not possess the necessary detection limits to quantify multiple As species at sub mg [kg.sup.-1] levels. Speciation studies of solids generally require extraction of the As from the solid and analysis of the resulting extracted solution. Ideal extraction techniques are those that extract all of the As from a particular sample without changing the speciation from that in the solid. Moreover, the ideal analytical system would identify all the As species in that extracted solution even at very low concentrations; currently, high-performance liquid chromatography-inductively coupled plasma-mass spectrometry (HPLC-ICP-MS) is closest to the ideal analytical system. Recently, dilute (1-2 %) extractant solutions of HN[O.sub.3] have been shown to give excellent recovery of As species from reference materials and from rice and rice products [22], while being simpler than earlier trifluoroacetic acid (TFA) extraction procedures [23].

Infants and young children may be particularly vulnerable to dietary sources of As. The 2009 European Food Safety Authority (EFSA) report found that dietary exposure to inorganic As for children under age three is ~2 to 3 times higher that of adults [24]. Many families introduce rice cereal as the first solid food for 4-6-month-old infants. These rice-based cereals are particularly high in As relative to other foodstuffs, and Meharg et al. [15] estimated a median intake of As of 0.21 [micro]g [kg.sup.-1] [d.sup.-1] for an average 20-lb, 1-year-old baby, by consumption of a single 20-g serving, exposure which exceeds that of an adult drinking water containing 10 [micro]g [l.sup.-1] As (0.17 [micro]g [kg.sup.-1] [d.sup.-1]). This latter adult exposure value is based on daily consumption of 11 of tap water as estimated by the EPA [25]. Exposure to As from consumption of rice cereal is high both because of the high concentration of As in rice cereal and the low body mass of a child.

Rice and derived products like starch, flour, and syrup are used to fortify a number of processed baby foods, including formula (powdered baby milk), jarred purees and strained foods, and snack items. In this study, we have determined total As and As speciation in a number of infant formulas and first foods. For formulas, we compared dairy- and soy-based products. For first foods, we tested fruit and vegetable purees, plus more complex stage 2/3 purees that contain meat and grains, and determined whether rice-containing and non-rice-containing and meat and vegetarian products differed in their As concentrations. The work was conducted in support of an ongoing birth cohort study focusing on environmental health effects of early life exposure to As.

MATERIALS AND METHODS

Infant formulas and first foods were purchased from supermarkets in the Hanover, New Hampshire area and were chosen from popular brands and to reflect the diversity of ingredients in these foods. Foods were analyzed directly from the sample container and were not further dried or homogenized. For total As analysis of infant formulas, approximately 250-mg samples were acid-digested using 2 ml 50:50 optima HN[O.sub.3]:[H.sub.2]O by microwave digestion (MARS XPRESS, CEM, Mathews, NC) with a 10 min ramp and 10 min hold at 180[degrees]C. The digested sample was then diluted with deionized (DI) water to a final volume of 10 ml. The diluted sample was analyzed for As by ICP-MS (7700x, Agilent, Santa Clara, CA) using He as a collision gas.

Purees and stage 2/3 foods were "open vessel"-digested in concentrated HN[O.sub.3]. Between 1-2 g of each product was weighed into a 50-ml polypropylene tube and 2 ml of acid was added. The vials were lightly capped and heated in a microwave at 95[degrees]C for 30 min. The samples were allowed to cool, 250 [micro]l of [H.sub.2][O.sub.2] was added, and the samples were taken through a second heating step. The samples were diluted to 10 ml and the weight was recorded. An aliquot of digested sample was then centrifuged at 13 300 rpm for 30 min, and a 1-ml aliquot for the supernatant was filtered (0.45 [micro]m) and diluted to 4 ml with DI water.

For speciation analysis, formula and food samples were extracted with 1 % HN[O.sub.3]. Approximately 2 g of sample was weighed into a 50-ml polypropylene tube and 20-40 ml of 1 % HN[O.sub.3] was added. The tubes were lightly shaken then taken through a progressive microwave heating program of 10 min at 55[degrees]C, 10 min at 75[degrees]C, and 30 min at 95[degrees]C. An aliquot was then centrifuged and/or filtered (0.45 [micro]m) prior to speciation analysis. The samples were further spin-filtered through 10 KDa spin filters (VWR, Radnor, PA) to remove larger molecular weight constituents that could foul the ion-exchange columns.

Speciation analysis was by anion-exchange chromatography coupled to ICP-MS. An Agilent LC1120 was used as the liquid chromatography system. Two different exchange columns were used, a Hamilton PRP X100 and a Dionex AS16 column. For the Hamilton column the eluant was 20 mM N[H.sub.4][H.sub.2]P[O.sub.4] at pH 6 and a flow rate of 1 ml [min.sup.-1]. For the Dionex AS 16 column, the gradient elution method reported in Jackson and Bertsch [26] was used with tetramethyl ammonium hydroxide as the mobile phase and a flow rate of 1 ml [min.sup.-1]. In both methods, the effluent was introduced directly to the ICP-MS equipped with a seaspray nebulizer (Glass Expansion, Pocasset, MA). The ICP-MS was operated as described above.

Inorganic As standards were obtained from Inorganic Ventures (Christiansburg, VA), while MMA and DMA were prepared from salts of monosodium methane arsonate (Chem Services, West Chester, PA, USA) and cacodylic acid (Sigma-Aldrich, St. Louis, MO), respectively. Calibration standards were prepared daily from serial dilution of stock species standards. Quality control for total digestion and analysis included triplicate analysis for all formula samples and duplicate and spike analysis at a frequency of one each per batch of 20 samples for the purees and stage 2/3 foods. Rice flour National Institute of Standards and Testing (NIST) Standard Reference Material (SRM) 1568a (Gaithersburg, MD) was used as a quality control material for both total As measurements and As speciation. Although As species are not certified for this SRM, reproducible consensus values have been demonstrated from many studies. We determined total As in NIST 1568a to be 318 [+ or -] 26 ng [g.sup.-1] (n = 8), the certified value being 290 [+ or -] 30 ng [g.sup.-1]. For As speciation, we determined DMA to be 200 [+ or -] 17 ng [g.sup.-1] and inorganic As to be 105 [+ or -] 17 ng [g.sup.-1], which are in the range reported by other studies [9].

We used mixed model, nested analysis of variance (ANOVA) (JMP version 8.0.2) to test whether mean total As concentrations differed with product formulation. For example, we compared formulas (1) with and without dairy and (2) with and without rice. For the purees, we compared fruit- vs. vegetable-based products, and (after noticing that pears were particularly high in As), pears vs. other kinds of fruits. In each model, we treated the factor of interest (dairy/soy, rice/not-rice, fruit/vegetable) as a fixed main effect, with a random effect of the product name (e.g., Brand A sweet potatoes) nested within that main effect to account for replicate measurements for most of the products tested. Finally, for the stage 2/3 foods, we compared products with and without rice and with and without meat, in a two-factor ANOVA, with product name nested within each rice x meat treatment. For the formulas, As concentrations met the assumptions for analysis without transformation, but for the purees and stage 2/3 foods, [log.sub.10]-transformation was required to homogenize variance.

We also make some estimates of average As exposure for infants through to 1-year-old babies based on personal experience of infant diets and feeding frequency and average of the 50th percentile weights for 3-, 6-, and 12-month-old children taken from the WHO child growth standards (http://www.who.int/childgrowth/standards/Chap_4.pdf). For comparison, we use an upper exposure metric suggested by Meharg [15] that a 60-kg adult consuming 1 l of drinking water at the EPA/WHO limit would consume 0.17 [micro]g As [kg.sup.-1] [d.sup.-1].

RESULTS AND DISCUSSION

Infant formulas

We analyzed 15 infant formulas comprising 5 main brands. The formulas were further classified as to whether they were dairy- or non-dairy-based, and whether they contained rice starch. Arsenic totals were then statistically evaluated between these classes for significant differences. Arsenic was detectable in all infant formulas, with values ranging from 2.2 to 12.6 ng [g.sup.-1] (Table 1). The mean As concentration was significantly lower in dairy-based formulas than those without dairy (nested ANOVA [F.sub.1,13] = 13.3, P = 0.003). Most (92.9 %) of the variability not explained by the fixed effect of dairy was explained by formula type; triplicate samples had coefficients of variation <16.9 %.

Arsenic speciation was evaluated on 1 % HN[O.sub.3] extracts for the 9 infant formulas with total As >6 ng [g.sup.-1]. The concentration of As species in the extracted formula samples were near the limit of detection of the IC-ICP-MS technique for either ion-exchange column. Nevertheless, the results from both ion-exchange columns were in general agreement: As speciation in the formulas was almost exclusively inorganic and the major As species was As(V); both ion chromatography columns showed that infant formula (IF) 11 had a higher proportion of As(III) than the other formulas. Low concentrations of DMA (~0.5 [micro]g [l.sup.-1]) were quantifiable only on the AS16 column. Recoveries for the Hamilton column (sum of species/total digested As concentration) ranged from 54 to 102 % with an average of 76 %, while for the As16 column they ranged from 72 to 145 % with an average of 80 %. Both columns had an As species eluting in the void volume for many of the infant formulas, which may be either arseno-betaine or other As species not retained by anion-exchange chromatography.

Few other studies have reported As levels in infant formulas. One study, which used a similar approach to that described here, reported that infant formulas ranged from 12 to 17 ng [g.sup.-1] and that the major species were inorganic As and DMA [27]. A more recent study reported As concentrations for formulas reconstituted per the manufacturer's instructions with DI water and these ranged from <1 to 1.6 [micro]g [l.sup.-1] [28]. Applying a similar calculation to our data, 1 scoop formula per 60 ml water and an average scoop weight of 9 g yields formula As concentrations of 0.3-1.8 [micro]g [l.sup.-1]. Hence, there is general agreement between studies on the concentration levels of As in main brand formulas. Using the As concentration range from Table 1, we calculate that the As exposure of a 3-month-old 6.2-kg infant consuming 6 120-ml bottles of formula daily, would be between 0.036-0.21 [micro]g As [kg.sup.-1] [d.sup.-1] solely from formula. This higher range value exceeds the 0.17 [micro]g [kg.sup.-1] [d.sup.-1] limit referred to earlier, of an adult drinking 11 of water at the WHO/EPA limit, and suggests the potential vulnerability of infants, because of their low body weight, to even ostensibly low concentrations of As in food.

Purees

Arsenic concentration was also analyzed in three different brands of fruit and vegetable purees (n = 40) targeted at 6-12-month-old infants (Table 2). For two brands (D, G) the total As concentration ranged from 0.32 to 7.8 ng [g.sup.-1]. For brand E, most purees were low in total As (1-4 ng [g.sup.-1] with a MDL = 0.15 ng [g.sup.-1]), except for the pear-containing products, which had an average As concentration of 16.6 ng [g.sup.-1]. Given the brand-specific nature of these high As pear products, these high concentrations are likely source related rather than a property of pears in general. Overall, fruit purees had marginally higher As concentrations than vegetable purees ([F.sub.1,40.7] = 3.86, P = 0.06), but this result was driven entirely by pear products from one brand; when pear-containing products were excluded from the analysis, fruit and vegetable purees were not different ([F.sub.1,34.01] = 0.12, P = 0.73). Within the fruit purees, pear-containing products had significantly more As ([F.sub.1,18.17] = 32.3, P < 0.0001). Our results are similar to those of Vela and Heitkemper [27] who report an As range of <1-24 ng [g.sup.-1] for infant puree food products and the prevalence of inorganic As in the speciated samples.

For the high As, pear-containing products, we measured As speciation using the Hamilton PRP X100. Arsenic speciation was very similar for each pear-containing product, with inorganic As > DMA. Inorganic As (as the sum of As(III) and As(V)) ranged from 76 to 83 %, and the overall recovery of As species (sum of species as a percent of the total As determined separately) ranged from 80 to 96 %.

Total As concentrations per serving for these fruit and vegetable purees ranged from 0.03 to 2.3 [micro]g. A typical 7-month-old infant ([approximately equal to]8 kg) consuming these products might eat 1.5 full jars daily. At the median exposure (0.25 [micro]g/serving), this infant would be exposed to [approximately equal to] 0.05 [micro]g As [kg.sup.-1] [d.sup.-1], or of the "safe" adult level derived from drinking water. However, if this infant were eating the median jar of pears daily (1.6 [micro]g As/serving), the exposure would be 0.2 [micro]g [kg.sup.-1] [d.sup.-1], above the safe adult level. Also, an infant of this age would still be formula fed, contributing more As to the daily exposure total.

Stage 2/3 foods

We also determined As in more complex infant foods containing multiple ingredients including meat, vegetable, and grain products; these foods are typically marketed at infants aged 9-15 months. N.B. no "rice only" foods, such as cereals, were considered in this study; previous studies have shown these to have high inorganic As concentrations with median values of 110 ng [g.sup.-1] [15]. We subclassified these foods into meat (M) or vegetarian (V), and rice- or non-rice-based. Total As ranged from below detection (ca. 1-3 ng [g.sup.-1] depending on the extent of dilution of the digested solid) to 22 ng [g.sup.-1] (Table 3). Statistically, there was an interaction between meat and rice content [F.sub.1,37.25] = 14.42, P = 0.0005): foods with both meat and rice had the highest As concentration. Foods without rice had the lowest mean total As concentrations (least-squares mean of 3.75 ng [g.sup.-1] for neither meat nor rice and 2.34 ng [g.sup.-1] for meat but not rice). Of the foods with rice, those without meat had lower As concentrations (least-squares mean of 11.90 ng [g.sup.-1]) than those with meat (least-squares mean of 18.45 ng [g.sup.-1]).

Arsenic speciation was determined in foods where As > 5 ng [g.sup.-1] and was again found to be predominantly (>70 %) inorganic As. This was somewhat unexpected given that for these foods the concentration of As is related to the presence of rice and that DMA is the major species in U.S. rice, although rice from other countries such as Bangladesh and India have higher ([greater than or equal to] 80 %) proportion of inorganic As [9]. Alternatively, the rice in these foods may have higher levels of rice bran, which is known to be higher in inorganic As than bulk grain [18], or there may be additional (non-rice) sources of inorganic As from other ingredients in these foods. We note that inorganic As is the major species (75-90 %) in rice products such as crackers, noodles, and puffed rice [19] and that our results for baby foods show similar levels of inorganic As.

Total As content per 170 g (brand G) or 113 g (brands D and E) serving of these foods ranged from 0.17 to 3.7 [micro]g, with a median of 1.3 [micro]g. If a 10-kg infant (~1 year old) consumed 3 full jars at the median As concentration each day, s/he would be exposed to 0.39 [micro]g As [kg.sup.-1] [d.sup.-1], more than twice the 0.17 [micro]g [kg.sup.-1] [d.sup.-1] safe adult As exposure level. Even at the lowest concentrations, the daily exposure would be 0.05 [micro]g [kg.sup.-1] [d.sup.-1] solely from these jarred foods, before any consideration of other potential As sources such as cereals or water.

CONCLUSIONS

Although we report relatively low concentrations of As (<1-23 ng [g.sup.-1]) in formulas, purees, and multiple- ingredient infant foods, these levels are potentially of concern because As is present mainly in the more toxic inorganic form. In addition, the low body weight of infants means that when expressed on a [micro]g [kg.sup.-1] [d.sup.-1] basis, even these low concentrations result in exposures that are greater than for an adult drinking water at the WHO/EPA safe drinking water level. Additionally, our results and theoretical dietary intakes do not take into account additional As present in water used to reconstitute the infant formulas, which can be significant in our geographical region (New England, USA) [29] and elsewhere. We also do not consider rice-based cereals, which can be an order of magnitude higher in As than the foods reported here [15].

It is clear that food is a significant route of As exposure for infants that must be considered in any epidemiological study. Although the United States has no regulations governing the As concentration of foodstuffs, China has set a level of 150 ng [g.sup.-1] inorganic As for rice [30]. However, in the case of infants, where the per kg exposure rate is so much higher, then even this limit would appear to be too high.

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Table 1 Total As concentrations, speciation, and formula type for 15 main brand formulas.

Total As (ng [g.sup.-1])	Dairy	Rice	Species recovery	% Inorganic As	Brand
5.36 [+ or -] 0.21	YES	NO	n.s.		A
11.27 [+ or -] 0.35	NO	NO	88.20%	100%	A
9.29 [+ or -] 0.43	NO	NO	88.61%	100%	A
11.89 [+ or -] 0.64	NO	YES	66.76%	100%	A
5.76 [+ or -] 0.4	YES	NO	n.s.		B
6.95 [+ or -] 0.43	NO	NO	102.84%	100%	B
11.43 [+ or -] 1.09	NO	NO	84.25%	100%	B
6.02 [+ or -] 0.26	YES	YES	n.s.		B
8.19 [+ or -] 0.63	YES	YES	54.48%	100%	B
8.14 [+ or -] 0.77	YES	NO	55.31%	100%	C
9.38 [+ or -] 0.31	YES	NO	62.75%	100%	D
2.92 [+ or -] 0.33	YES	NO	n.s.		D
9.62 [+ or -] 1.35	NO	NO	77.58%	100%	E
3.42 [+ or -] 0.2	YES	NO	n.s.		F
2.6 [+ or -] 0.44	YES	NO	n.s.		F

n.s. = not speciated.

Table 2 Total As concentration in 41 first-food purees.

Ingredients	As (ng [g.sup.-1])	Brand
Pears and mango	15.01	E
Sweet potatoes	1.45	E

Apples and apricots	1.48	E
Winter squash	0.68	E
Sweet potatoes	2.78	E
Apples and blueberries	0.93	E
Apples	0.69	E
Prunes and oatmeal	1.74	E
First prunes	1.25	E
Apples and plums	0.97	E
Pears	13.55	E
Pears and raspberries	20.20	E
Carrots	1.68	E
Peas	3.14	E
Squash	1.90	E
Corn and butternut squash	0.48	E
Pears	17.52	E
Apples	2.51	E
Bananas	3.99	D
Pears and wild blueberries	1.00	D
Sweet potatoes	5.03	D
Apples	2.47	D
Green beans	3.21	D
Pears	4.64	D
Carrots	1.75	D
Squash	1.14	D
Apple and strawberries	1.2	D
Apples	6.74	D
Peaches	3.34	D
Prunes	2.01	D
Sweet peas	1.06	D
Select prunes	1.63	D
Select sweet potatoes	7.81	D
Green beans	0.9	G
Squash	0.48	G
Pears	3.17	G
Sweet peas	0.74	G
Sweet potatoes	4.28	G
Sweet carrots	2.07	G
Bananas	0.32	G
Applesauce	0.65	G

Table 3 As concentration and selected speciation in second- and third-stage foods.

Food type	Total As (ng [g.sup.-1])	Rice	Species recovery
Meat and veg	<3.4	NO	n.s.
Meat and fruit	4.41	NO	n.s.
Meat, rice, and veg	22.34	YES	74.28 %
Meat and veg	12.52	YES	n.s.
Meat and pasta	18.32	YES	79.19 %
Veg and rice	10.33	YES	91.18 %
2 veg	6.29	YES	133.04 %
Rice and pulses	9.61	YES	92.67 %
Veg medley	11.28	YES	82.12 %
Meat and fruit	18.84	YES	88.56 %
Meat and fruit	13.42	YES	67.30 %
Meat and broth	13.81	YES	75.33 %
Meat and rice	14.15	YES	110.29 %
Fruit and oatmeal	1.74	NO	n.s.
Fruit and rice	17.8	YES	98.35 %
Meat and broth	<3.4	NO	n.s.
Meat and broth	5.43	NO	n.s.
Meat and broth	<3.4	NO	n.s.

Food type	% Inorganic As	Brand
Meat and veg		D
Meat and fruit		D
Meat, rice, and veg	72.1 %	D
Meat and veg		D
Meat and pasta	77.9 %	D
Veg and rice	87.4 %	E
2 veg	90.8 %	E

Rice and pulses	83.7 %	E
Veg medley	85.4 %	E
Meat and fruit	89.8 %	E
Meat and fruit	87.5 %	E
Meat and broth	86.2 %	E
Meat and rice	75.2 %	E
Fruit and oatmeal		E
Fruit and rice	77.2 %	E
Meat and broth		G
Meat and broth		G
Meat and broth		G

n.s. = not speciated.