Avoiding Rice-Based Cadmium and Inorganic Arsenic in Infant Diets Through Selection of Products Low in Concentration of These Contaminants

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Abstract
Cadmium in the diet is of concern as it is a renal toxicant and a carcinogen, with a half-life in the body measured in decades. Inorganic arsenic is a chronic carcinogen. For many subpopulations, rice and rice products may be the dominate source of cadmium and inorganic arsenic. In particular, rice porridge, cereal and cake are widely used to feed infants (children < 4.5 years old). In the EU standards for cadmium infant foods in general has been set at 40 μg/kg w.wt., and for inorganic arsenic in rice-based infant foods the standard is 100 μg/kg w.wt.. Here we report cadmium and inorganic arsenic concentrations in rice products marketed for infants, and rice containing products that infants may eat but that are not specifically designated for infants. It was found that while rice-based infant foods conformed to the standards, their non-infant food (generic) analogues did not. Non-infant rice crackers and puffed rice cereals, in particular, had concentrations above these standards for both cadmium and inorganic arsenic. Polished pure rice grain purchased in the UK, but sourced from different countries, was also problematic. Basmati, Italian, Spanish and Thai rice, either exceeded one or the other of the cadmium and inorganic arsenic safety thresholds for infants, or both. Egyptian rice grain was particularly low for both toxins. Therefore, if those responsible for infants want to lower exposure to cadmium and inorganic arsenic, they should stick to foods specifically labeled for infants, or carefully source low cadmium and inorganic arsenic rice-based products that are not specifically labeled as being for infant consumption, or minimize exposure to rice-based foods.

Keywords Arsenic · Cadmium · Exposure · Infants · Rice

Introduction

Human exposure to the carcinogens cadmium and inorganic arsenic, in non-smokers, is primarily through diet (EFSA 2009, 2012; BrF 2018). In particular, rice, the grain staple of half-the-world, tends to be elevated in these toxicants (EFSA 2009, 2012; BrF 2018). For subsistence, rice diets elevated exposure to both cadmium (Meharg et al. 2013; Pastorelli et al. 2018) and inorganic arsenic (Meharg et al. 2009) has given rise to international concern. Globally, most problematic is the exposure of infants to cadmium and inorganic arsenic as they are particularly susceptible to these elements: elevated early life exposure to cadmium and inorganic arsenic are thought to lead to poorer lifetime health outcomes (Carey et al. 2018; Gardener et al. 2019; Ljung et al. 2011; Meharg et al. 2008a, b). Unfortunately, rice is widely fed to infants as it is gluten free, and is often used for weaning (Meharg et al. 2008a, b). Exacerbating exposure to children is the fact that infants consume circa. 3 times more food on a bodyweight basis than adults (EFSA 2009).

Standards have been set in the EU in recognition of the fact that cadmium (European Commission 2006) and inorganic arsenic are problematic in the food-chain (European Commission 2015). Cadmium standards are set for a wide range of food items (European Commission 2006). Of relevance to rice is the fact that maximum concentrations allowed in polished (white) rice grain for cadmium and inorganic arsenic are both 200 μg/kg w.wt. (European Commission 2006, 2015). Infant standards for cadmium (European Commission 2006) and inorganic arsenic (European Commission 2015) have been set lower, at 40 and 100 μg/kg.
w.w.t., respectively. However, the inorganic arsenic standard is set on the rice used in formulation, regardless of final concentration, and cadmium is set on absolute concentrations in the product. 

As products not specifically labeled as being for infant consumption are fed to infants, there is a gap in the legislation. If parents want to feed children non-infant rice-based foods low in inorganic arsenic and cadmium, advice needs to be given as to what food items are suitable. Knowledge of what potential products are high or low in these toxicants is complicated by the fact that the geographic origin of rice effects both cadmium (Meharg et al. 2013; Shi et al. 2019) and inorganic arsenic (Meharg et al. 2009; Carey et al. 2020) content. Here we report cadmium inorganic arsenic concentrations in rice-based products available on the UK market, examining in particular in how non-infant food relates to that for infants, including rice grain by geographic origin, so that informed choices can be made by those responsible for the diets of children.

**Materials and methods**

Rice containing baby products were sampled in 2018 from supermarkets in Northern Ireland to reflect consumer choice. Inorganic arsenic, but not cadmium, was previously reported in these samples (Carey et al. 2018). Survey was systematic in that 3 stores of 6 major retailers (ASDA, Boots, Holland and Barrett, Marks and Spencer, Sainsbury’s, Tesco) were visited. All rice-based baby foods, crackers and cereals available at the time of survey in each store were sampled. These included baby rice, rice cereal and rice cake. Details of brand, purchase date, store details, ingredients, and country of origin were recorded and reported in Carey et al. (2018). Note that while sampling took place in Northern Ireland, the source of the rice, brands and distributors are international, and commonplace throughout the UK and EU.

Additionally, polished rice grain were sampled. They were obtained from supermarkets, health food shops and specialty shops in Northern Ireland, with the same sampling strategy as for baby food products. Samples were collected during 2013–2018. Wholegrain is less common and also is not normally fed to young infants and is, therefore, excluded. Grains represented 6-regions, 5 commonly available: basmati (from India & Pakistan), Spanish, Italian, Thai and the USA. Specialty Egyptian rice was purchased from Halal shops. The polished rice grain data were abstracted from a wider global survey of inorganic arsenic and cadmium in rice reported by Carey et al. (2020) and Shi et al. (2019), respectively, where sampling strategies and experimental design are outlined in full. Sampling frequencies are shown in relevant figures where individual data points are plotted. While the grain survey was opportunistic, it follows the pattern used in similar surveys (Shi et al. 2019). As long as the surveys are interpreted with suitable caution, they provide valuable information, as discussed by Shi et al. (2019).

The samples were dried in a Christ LD freeze dryer, powdered on a rotary ball mill (Retch PM 100 planetary ball mill) using a zirconium oxide—lined grinding chamber and 20 mm zirconium oxide—plated marbles. Powdered samples were then weighed, ~100 mg, accurately, using discovery OHAUS digital weighing scales into labeled 50 ml polypropylene (pp) centrifuge tubes (VWR, D&H and similar). A rice flour certified reference material (CRM), NIST 2018, was also used for each analysis batch, with a batch consisting of 40 samples, and each batch also included an analytical blank.

For cadmium analysis, to each centrifuge tube 2mls of BDH Prolabo Aristar 69% nitric acid was added, including the blanks. Tubes were vortexed briefly and left overnight to soak. Following this period, 2mls of BDH Prolabo Analar Normapur 30% hydrogen peroxide was added to each centrifuge tube via pipette. Tubes were then left open for 15 min to outgas. Tubes were then placed into the carousel of a CEM Mars 6 1800 W microwave digestor, and the appropriate digestion program selected. The microwave program heated the samples up to 95 °C gradually through a 3-stage process over a period of 35 min, then digested the samples at 95 °C for 30 min. After cooling, a rhodium internal standard (Fluka Analytical) was added, to give a final concentration of 10 µg/l, to each sample, and the tubes were made up to their final weights (~ 30 g) with deionized water, with precise weights recorded. Multi-Element 2 (SPEX CLMS-2 Multi-Element Solution 2, matrix: 5% HNO₃) and Multi-Element 4 (SPEX CLMS-4 Multi-Element Solution 4, matrix: water/Tr-HF) were used to make up all standards in a range of 0–100 µg/l. The standard tubes were then made up to final weight (50 g) with 1% HNO₃. For analysis, 10 ml from the final digestate was poured into 15 ml polypropylene tubes (VWR) and placed into the auto sampler rack (Cetak ASX-520 Auto Sampler) in a predetermined random run order.

For arsenic speciation, 10 ml of 1% conc. Aristar nitric acid was added to the ~0.1 g of powdered rice and allowed to sit overnight. Samples were then microwaved for 30 min at 95 °C using a 3-stage heating program: to 55 °C in 5 min held for 10 min., to 75 °C in 5 min, held for 10 min to 95 °C in 5 min, held for 30 min. On cooling the digestate was diluted to 10 ml with deionized distilled water and then centrifuged for 15 min at 4500 rpm for. To a 1 ml aliquot in a 2 ml polypropylene vial 10 µl of analytical grade H₂O₂ was added, converting any arsenite to arsenate, facilitating chromatographic detection.

Shi et al. (2020) outlines the cadmium analytical protocols used here. Summarizing, for cadmium quantification samples was analyzed by ICP-MS (Thermo Scientific iCap Q ICP-MS), interfaced with an auto-sampler. The
ICP-MS operating conditions were: forward RF power-1550 W; nebulizer gas flow~ 1 l/min, nebulizer sample flow rate~ 0.35 ml/min. Helium was used as a collision gas at a flow rate of 5 ml/min. Five cadmium standards were made up including one blank, all in 1% HNO₃. Cadmium recovery was, mean ± SE, 105 ± 3.2%. The certified concentration of NIST rice flour CRM was 24 μg/kg d.wt. The limit of detection (LoD) for cadmium was 0.5 μg/kg d.wt. for infant foods, but at 9.8 μg/kg d.wt. for rice grain.

For inorganic arsenic quantification analytical details are given in Carey et al. (2018, 2020). Summarizing, sample solutions were analyzed using a Thermo Scientific IC5000 Ion Chromatography (IC) system fitted with a Thermo AS7, 2×250 mm column (and a Thermo AG7, 2×50 mm guard column), using a gradient mobile phase (20 mM to 200 mM ammonium carbonate linearly over 15 min., using a flow rate of 0.3 ml/min), interfaced with a Thermo ICAP Q ICP-MS that monitored m/z+ 75, using helium gas in collision cell mode. Calibration was quantified conducted using a dimethylarsinic acid (DMA) calibration series, and compounds identified through the use of authentic arsenobetaine, arsenate, DMA and monomethylarsonic (MMA) standards. Recovery of the NIST rice flour CRM for inorganic arsenic (n = 76) was 99 ± 1.1%, with a limit of detection of 3 μg/kg d.wt..

For infant foods and their generic equivalents, the means of the 3-replicates were obtained. Subsequently, all intercomparisons, between product type and between regions, where appropriate, were conducted using non-parametric tests as the cadmium and arsenic data were not normally distributed using GraphPad Prism for Mac, v. 8. If a sample was below LoD, half-LoD was used in statistical analysis.

Rice grain is normally dried to 12% for storage (University of Arkansas, 2020), and this conversion factor can be used to approximate wet weights from dry weights if required for grains. This ~12% moisture lost on drying in effect means that d.wts. need to be multiplied by 1.12 to convert to w.wt., and this should be born in mind when interpreting data.

## Results

The cadmium and inorganic arsenic concentrations in rice-based infant foods and their non-infant food analogues, in relationship to the EU regulations pertaining to infant foods, are shown in Fig. 1. For cadmium, all infant foods were below the EU standard for processed infant foods of ~44.8 μg/kg d.wt., which equates to 40 μg/kg w.wt.. Indeed, all but 4 samples were below 20 μg/kg d.wt.. Mixed cereal cakes had the highest median at 12.8 μg/kg d.wt., and the lowest was mixed grain porridge at 5.1 μg/kg d.wt. These infant products significantly differed (P = 0.019). Not labeled for infants rice-based foods, in contrast, had 6 out of 44 samples exceeding ~44.8 μg/kg d.wt. cadmium, 2 of each from mixed cereal cakes, pure rice cakes and pure rice breakfast cereals, with those exceeding the standard ranging from 46.0 to 66.2 μg/kg d.wt.. For cadmium there was no significant difference (P > 0.05) between non-infant products, and for all products.

For inorganic arsenic a proxy standard of 100 μg/kg w.wt., equating to ~112 μg/kg d.wt., was used to compare results to EU guidelines. This standard is denoted as “proxy” as the EU inorganic arsenic standard is set on raw material grain used to produce the products (European Commission 2015). All infant foods were below the ~112 μg/kg d.wt. proxy standard (Fig. 1). Non-infant products had some samples above the proxy standard, with pure rice cakes and pure rice cereals both having medians above ~112 μg/kg d.wt..
All infant foods were below the EU threshold (Fig. 1), but mixed grain infant cereals (7.8 µg/kg d.wt.) and porridges (11.9 µg/kg d.wt.) had medians considerably below mixed cake (60.6 µg/kg d.wt.), and pure porridge (65.9 µg/kg d.wt.) and cake (74.4 µg/kg d.wt.). There were highly significant differences ($P < 0.0001$) when comparing all products, and within infant products. Differences between non-infant products were significant with $P = 0.014$.

Plotting individual products cadmium and inorganic arsenic non-infant pure rice cakes had 2 products that exceeded both infant (proxy) inorganic arsenic and cadmium standards (Fig. 2). Pure rice cereals borders on exceedance of both standards for 3 products, and two mixed grain rice cakes are in a similar position. Linear regression of inorganic arsenic plotted against cadmium for the entire data set was significant ($P < 0.0001$). The slope, origin set to zero, was 0.031, and thus inorganic concentrations are ~threefold higher than cadmium in rice-based baby food products.

When considering pure polished rice grains the median cadmium content of basmati rice was 33.4 µg/kg d.wt., with a 75th percentile of 55.4 µg/kg d.wt.. This was above the infant rice standard of ~44.8 µg/kg d.wt. (Fig. 3). Italian rice had a median of 40.1 µg/kg d.wt., with a 75th percentile of 60.2 µg/kg d.wt.. The maximum for Italy was 117 µg/kg d.wt. Thai rice had a 75th percentile above the EU estimated d.wt. threshold, at ~44.8 µg/kg d.wt.. Spain had a low median and 75th percentile, but the maximum value was above the EU threshold. Egyptian and Spanish rice had medians at the half-LoD (4.9 µg/kg d.wt.), and the USA double this, though with a low sample size ($n = 3$). The medians between countries differed significantly when a Kruskal–Wallis analysis was performed on the data ($P < 0.0001$).

Italian rice had the highest median inorganic arsenic at 106 µg/kg d.wt., with the d.wt. corrected standard being ~112 µg/kg d.wt. (Fig. 3). The 75th percentile was 126 µg/kg d.wt., and the maximum 244 µg/kg d.wt. Basmati (28.9 µg/kg d.wt.) and Egyptian (30.2 µg/kg d.wt.) had the lowest medians, with maximums well below EU d.wt. approximated threshold of ~112 µg/kg d.wt.. Thailand and USA also had maximums below the EU threshold. Medians were highly significantly different between regions ($P < 0.0001$). The maximum Spanish sample was 210 µg/kg d.wt., but median and 75th percentile were well below the ~112 µg/kg w.wt. standard.
Considering cadmium and inorganic arsenic together, it is only Egyptian rice that is low in both toxins. Italian rice was problematic for both toxicants, while Spain, Thailand and USA were intermediate. When inorganic arsenic is plotted against cadmium concentrations (Fig. 4), Italian rice is the only region where medians exceed both d.wt. corrected limits, while Egyptian rice is the only one to exceed none. Linear regression of the entire dataset, with origin set to zero, was significant ($P < 0.0001$). The slope of inorganic arsenic verses cadmium content was 0.27, similar to the corresponding slope for the similar plot for processed infant foods (Fig. 2).

**Discussion**

From the findings reported here, rice products not labeled as being specifically for infant consumption are problematic with respect to their cadmium and inorganic arsenic contents if they are to be used as infant foods. This is because rice products not specifically labeled for infant consumption routinely exceed the standards for either cadmium or proxy inorganic arsenic set for infant foods, or both, in a non-predictable manner. The only exception to this statement is Egyptian rice, which is consistently low in both inorganic arsenic and cadmium. Puffed rice cereals are often marketed at children, with cartoon characters often on packaging. Puffed rice has a higher standard (300 μg/kg w.wt.) for inorganic arsenic than polished rice or rice destined for infant foods (European Commission 2015). Many rice-based products are formulated using puffed rice. It is confusing to set much lower standards for infant foods if infants are also exposed to generic foods that have higher standards? Rice milk tends to be elevated in inorganic arsenic (Meharg et al. 2008a, b). The UK Food Standards Agency (FSA) has asked that rice milk cartons have a warning printed on them that infants should not consume this product (FSA 2018). This UK rice milk arsenic approach shows that advice can be given clearly around toxicants to those providing foods for infants (FSA 2018). This labeling approach needs to be widened to all rice-based products, such as to puffed rice and polished rice grain, for both inorganic arsenic and for cadmium, if infants may routinely consume them.

The rice grain most suitable for infant consumption with respect to low cadmium and inorganic arsenic, available to European markets, is Egyptian. However, Egyptian rice is a specialty product, and on a global scale is relatively limited in availability, accounting for 0.8% of the global production, with only ~1% exported (FAO 2018). We have also identified that East African, Malawian and Tanzanian, rice is lower than Egyptian in both inorganic arsenic (Carey et al. 2020) and cadmium (Shi et al. 2019). Again, production from these two regions is limited on a global scale, accounting for ~0.5% of global production, with little export (FAO 2018). Also, there are agronomic constraints to expanding both East African (Meertens 2003) and Egyptian (Fan et al. 1997) rice production.

The lack of wide availability of low-arsenic and cadmium rice, and of labeling so that such products can be identified, then poses the question as to what can careers do to reduce infant exposure to these chemicals? One obvious solution is to avoid rice. Rice is preferred for a number of diets compared to gluten containing grains such as wheat and barley. Also, wheat, barley and oats can be high in cadmium (BfR 2018; Clemens et al. 2013; John et al. 1972).

Gluten free grains and grains are obvious alternatives to rice, such as maize, amaranth, quinoa, chia etc. This has been realized by infant food manufacturers producing...
products were rice is mixed with other grains for low inorganic arsenic (Carey et al. 2018). We show here that this is also effective for cadmium.

If polished rice grain is to be used, simple cooking techniques, presoaking rice and then cooking a large water:rice ratio, can be used to lower inorganic arsenic content, by 70–80% (Carey et al. 2015; Raab et al. 2009). Cadmium is not readily removed, so selection of low cadmium rice becomes particularly important. Egyptian and Spanish rice are relatively low in cadmium, as reported here. Wholegrain rice should be avoided if the aim is to minimize cadmium and inorganic arsenic in the diets of children as wholegrain has ~ double the inorganic arsenic content of polished rice (Meharg et al. 2009).

Concluding, in the absence of clear and comprehensive standard setting and governmental advice it is possible for those responsible for the diets of infants to simultaneously lower dietary exposure to inorganic arsenic and cadmium. This can be done by: choosing foods that are specifically designated only for infants (at least in the EU), avoidance of rice-based products, use foods specifically reduced in rice content, sourcing of rice grain that is low in both cadmium and inorganic arsenic, and cooking out inorganic arsenic out of low cadmium rice.

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