

Arsenic in the U.S. Food Supply

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Arsenic is a non-threshold carcinogen that causes bladder, lung and skin cancer. Worldwide, hundreds of millions of people in over 70 countries are exposed to arsenic through their drinking water. The highest concentrations have been found in South Asia where it is a major public health problem, particularly in areas of Bangladesh, Vietnam, and Cambodia. Epidemiological studies of drinking water exposures from this region (primarily Taiwan) formed the scientific basis for the EPA to reduce the safe drinking water limit of arsenic from 50 to 10 $\mu\text{g/L}$ in 2001.

Recently, attention has focused on human exposure to arsenic from food. A number of high profile media reports such as the Dr. Oz TV show and *Consumer Reports* have focused public attention on the concentration of arsenic in fruit juices and rice. There are no regulations for arsenic in food, however; in the case of apple juice, the FDA has recently proposed a guideline level for inorganic arsenic of 10 $\mu\text{g/L}$, bringing it in line with standards required for bottled water. For food, the setting of safe arsenic limits is complicated by a number of issues:

1. In food, arsenic can exist as different chemical species that differ in their toxicities with some of them even being harmless.
2. Arsenic chemical species vary between and even within foods, for example different types of rice.
3. Not all arsenic in food may be bioavailable upon ingestion – some passes through the gut unchanged.
4. Regulatory limits that were originally developed to prevent arsenic exposure via drinking water assume long term (decades) of daily exposure. This is clearly a reasonable assumption for water, but not for food, where food choice (and therefore arsenic exposure) is much more variable. Nevertheless, daily intake limits are ultimately expressed as micrograms of inorganic arsenic per kg bodyweight, and as such should be equally applicable to exposure via food.

Arsenic, agriculture, and animal farming have been inextricably linked throughout the 20th Century. Calcium arsenate and lead arsenate were used as pesticides in orchards prior to being phased out in the 1960s. However, organic arsenicals, such as dimethylarsenate (DMA) and monomethylarsenate (MMA) and their sodium salts, continue to be used as insecticides and herbicides in cotton farming, turf management, and on roadside grass verges, although they as well have now been mostly banned from use. It is also surprising to many people that poultry, turkey, and swine in the U.S. (and overseas in China, for example) are fed arsenic-containing compounds to promote growth as are chickens in order to control diseases such as coccidiosis.

In 1994, I was involved in a four-year project to study mixtures of coal fly ash and organic byproducts as soil amendments. At that time, I found it completely surprising that poultry manure was actually a much higher source of soluble arsenic than the fly ash. A little research soon revealed that several organic arsenic compounds were regulated for use as feed additives in the poultry industry. The main compound used in the US was 3-nitrophenylarsonic acid, roxarsone. The industry dogma at the time was that the compound was hardly metabolized by the chicken, and was excreted unchanged into the manure. Indeed, further studies showed that arsenic was present at high concentrations in the manure, up to 40 mg/kg, which is the maximum limit for sewage sludge to be applied to agricultural soils (although these EPA regulations didn't apply to animal manures). We also found both roxarsone and inorganic arsenic in manures, proving that these organic compounds were mineralized to the more toxic inorganic species. When the manures were mixed with soil, the arsenic species were very soluble and, presumably, available for plant uptake.

Other research groups investigated whether inorganic arsenic was taken up into the poultry meat. In 2006, the Institute for Agriculture and Trade Policy, a non-profit organization partly focused on sustainable agricultural practices, published "Playing Chicken: Avoiding Arsenic in Your Meat," which surveyed the concentration of arsenic in many common fast-food chicken meals; it found a range of concentrations from below detection up to around 50 ng/g (ppb), suggesting that some of the arsenic from the feed was actually making it into the meat. In 2011, the FDA conducted a controlled feeding study that clearly showed that increased inorganic arsenic was found in the meat and livers of roxarsone-fed chickens, even when they were slaughtered 5 days after cessation of the arsenic feed additive. The FDA and Pfizer reached an agreement to withdraw the use of roxarsone feed in poultry farming; now all of these feed additives, except nitrasone for turkeys, are banned.

Currently, there is a lot of interest in arsenic in rice. Scientific studies over the past 15 years, led primarily by the research group of Andrew Meharg and his colleagues in the U.K., have detailed the extent, amount, and speciation of arsenic in rice grain and rice products. They also elucidated the mechanism for arsenic uptake, which turns out to be an unfortunate consequence of paddy agriculture: the formation of soluble inorganic arsenic as arsenous acid, the need for rice to take up high concentrations of silicic acid, and the inability of the transporters to distinguish between the chemically similar silicic and arsenous acids. This means that compared to other grains such as wheat, or other staple carbohydrates, such as potatoes, rice has higher levels of arsenic. The actual concentration in rice is highly variable and driven mostly by the

type (cultivar) of rice, but is also influenced by other conditions such as the soil arsenic level. As suggested, rice grown without flooding accumulates much less arsenic. In the absence of any regulation removing the highest arsenic-containing rice grains or products, it is possible for people on rice-based diets to be consuming levels of arsenic comparable to that from drinking water at the safe drinking water limit.

The Dartmouth Superfund Program and Dartmouth's Children's Center for Environmental Health and Disease Prevention have been studying the effects of early life exposures to low levels of arsenic. As part of these studies, my group has analyzed many main brand infant and toddler formulas, first foods, such as purees, and second foods. Infant formulas are typically low in arsenic; however, two toddler formulas we tested, which used brown rice syrup as the main sugar substitute, had much higher levels of arsenic, with inorganic arsenic at or above the drinking water limit because the rice syrup contains arsenic originally present in the grain. The manufacturer of these products states that they have now modified their procedure to reduce the levels of arsenic in these products. The fruit purees we tested were low with a couple of exceptions; for example, we found some pear purees had arsenic levels of 20 ng/g, suggesting that, as with fruit juices, the source of the fruit rather than a trait of the fruit itself for accumulation leads to elevated levels. Not surprisingly, second foods that contained rice had significantly higher values than those that did not.

The FDA has also conducted extensive testing of rice and rice products, and has made the data available at <http://www.fda.gov/food/foodborneillnesscontaminants/metals/ucm319870.htm>. The data confirm previously published results; arsenic concentration is highly variable across rice types; brown rice tends to be higher in total arsenic and inorganic arsenic than white rice. The inorganic arsenic exposure per serving of rice ranges from < 2 to > 7 µg. All this research and media attention on arsenic in rice over the past decade has prompted organizations such as the World Health Organization (WHO), the European Food Safety Authority (EFSA), and now the FDA, to conduct expert meetings on arsenic exposure through food, particularly rice, with the goal of providing limits for consumer protection. In July 2014, the WHO Codex Alimentarius proposed a maximum level of 0.2 µg/g for inorganic As in polished rice.

This report is based on a talk that was given on May 14, 2014, at the NESACS Southeastern Massachusetts Area meeting at the Woods Hole Oceanographic Institution.