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Inorganic arsenic in starchy roots, tubers, and plantain and assessment of cancer risk of sub-Saharan African populations

Tuanwei Chen [a], Lihan Huang [b], Guoxin Lai [c], Guoying Chen [b,*]

[a] College of Food Science, Fujian Agriculture and Forestry University, Fuzhou, Fujian 350002, China
[b] U.S. Department of Agriculture, Agricultural Research Service, Eastern Regional Research Center, 600 E. Mermaid Lane, Wyndmoor, PA 19038, USA
[c] Department of Chemical Engineering, Meizhouwan Vocational Technology College, Putian, Fujian 351254, China

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ABSTRACT

Starchy roots, tubers, and plantain (RTP) are the staple food in sub-Saharan Africa (SSA), and also important energy sources in Asia, Europe, and America. In this work, inorganic arsenic (iAs) in these crops was separated and enriched by solid phase extraction (SPE), and quantified by hydride generation-atomic fluorescence spectrometry (HG-AFS). Overall, iAs in these crops ranged from 0.9 to 14.1 ng g^{-1} wet weight. Long-term cancer risk associated with iAs intake from these crops was assessed by Monte Carlo simulation based on iAs concentrations and historical consumption and population data. For 19 high RTP consuming SSA countries, life-time cancer risk was low with a mean target risk at 6.3×10^{-5} and a margin of exposure at 72.

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1. Introduction

Arsenic (As) is notorious in human history as the top poison and a ubiquitous environmental contaminant. For human, water and food are recognized as the top pathways of arsenic exposure. Unlike foods of animal origin, arsenic enters terrestrial plants from irrigation water and soil (Da Sacco, Baldassarre, & Masotti, 2013), and predominately exists in more toxic inorganic forms (Quaghebeur, Rengel, & Smirk, 2003; Schoof et al., 1999). High arsenic presence is indicative of contamination in local environment of either geological or anthropogenic origin. Such data are useful in evaluating arsenic contamination (Castro-Larragoitia, Kramar, & Puchelt, 1997) and planning long-term remediation. The International Agency for Research on Cancer (IARC) identified arsenic and arsenic compounds as Group 1 human carcinogen in 1987 (IARC, 1987). Inorganic arsenic (iAs) species are far more toxic than their organic counterparts; it causes not only cancers (Smith et al., 1992), but also a range of other adverse health effects including skin lesions (McDonald, Hoque, Huda, & Cherry, 2007), development toxicity (Rahman et al., 2007), neurotoxicity (Otto et al., 2007), cardiovascular diseases (Ana Navas-Acien et al., 2005), and diabetes (A. Navas-Acien et al., 2006). In 2010, the Food and Agriculture Organization/World Health Organization (FAO/WHO) set 3.0 μg kg bw^{-1} D^{-1} as the iAs lower limit on the benchmark dose for a 0.5% increased incidence of lung cancer ($BMDL_{0.5}$) (FAO/WHO, 2010).

In contrast to seafood that contains the highest total arsenic (tAs) among all food categories but mostly in less toxic or even innocuous organic forms (Edmonds & Francesconi, 1993), rice grows in flooded soil under anaerobic conditions and therefore contains higher iAs than other terrestrial plants, and much higher than foods of animal origin. Rice, together with rice products, has become the focus of regulatory efforts worldwide (Carbonell-Barrachina et al., 2012; Pasias, Thomaidis, & Piperaki, 2013). China set acceptable iAs level in rice at 200 ng g^{-1}, and the Codex Alimentarius Committee on Contaminants in Food proposed 200 and 300 ng g^{-1} draft iAs MLs in polished and raw rice, respectively (FAO/WHO, 2012).

* Corresponding author. Tel.: +1 215 233 6495; fax: +1 215 233 6642.
E-mail address: guoying.chen@ars.usda.gov (G. Chen).

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In comparison, arsenic in other terrestrial plants received much less attention except in contaminated area (Alam, Snow, & Tanaka, 2003). Starchy roots, tubers, and plantain (RTP) are prominent crops in a global scale reaching 849 million tonnes in 2012 (Table 1); 55% is traditionally used as food, the others as feed or for beer production (FAO Statistics Division, 2014). RTP crops plus fruits and vegetables account for 11% of dietary energy input for human, surpassed only by grains and animal products (FAO Statistical Division, 2008). Though traditionally known as roots and tubers, from botanical point of view tubers include root and stem tubers and cover cassava (*Manihot esculenta*), sweet potato (*Ipomoeu Batatas*), potato (*Solanales*), yams (*Dioscorea*), and edible aroids or cocoyams (*Colocasia esculenta* and *Xanthosoma*). RTP crops traditionally serve as food staples in sub-Saharan Africa (SSA) (Scott, Rosegrant, & Ringler, 2000). Plantain (*Musa* × *paradisiaca*) is among the major staple food crops in central Africa (Dury, Bricas, Tchango-Tchango, Temple, & Bikoi, 2002). Flours and starches derived from these crops also play important and unique roles in diet and culinary traditions among tropical populations, especially in Latin America, SSA, and South and East Asia. They are also used as the main ingredients in a great variety of secondary products such as bread, noodles, cakes, and cookies.

The objective of this work is to provide the first comprehensive survey of iAs exposure from RTP crops for high consuming SSA populations, based on which cancer risk was assessed by Monte Carlo simulation. Low-level presence of arsenic in these crops, however, demands reliable cleanup and extremely high detection sensitivity. Currently, both high performance liquid chromatography (HPLC)-inductively coupled plasma-mass spectrometry (ICP-MS) and hydride generation-atomic fluorescence spectrometry (HG-AFS) can fulfil this task. The latter, developed in this laboratory (Chen & Chen, 2014), focuses not on complete speciation but on inorganic arsenic which closely correlates to health implications; therefore it was ideal to apply to this survey after reliable cleanup by solid phase extraction (SPE).

2. Materials and methods

2.1. Samples collection and pretreatment

The samples, raw, dry, or flour, were purchased from local food stores in the Philadelphia area, PA, USA, or collected from ethnic stores, internet websites, even farmers or foreign sources.

Raw samples were skinned and cut into roughly 3 × 3 × 25 mm strings. Then, about 20 g samples and flours were placed in thin layers in Pyrex jars of known weights. The loaded jars were weighed, followed by oven dehydration at 80 °C overnight. The jars were weighed next day in 1-h intervals until constant weight was reached. Dried samples were then processed using a small mill (Depose 203, Krups, Mexico) into fine powders. Finally, after sealed tightly in glass containers, the powders were stored in a desiccator under room temperature.

Table 1
World production of roots and tubers in 2012 (FAO Statistics Division, 2014).

Crops	2012 production (T)
Cassava	269,125,963
Sweet potatoes	108,004,174
Potatoes	365,365,367
Yams	59,519,949
Aroids (Cocoyams)	9,988,552
Bananas[a]	36,600,427
Total	848,604,432

[a] Including plantain and fruit banana.

2.2. Analytical methods

The analytical protocol of this work was published previously (Chen & Chen, 2014), so only a brief description is provided here. Sample flours were digested in an oxidizing acidic medium using a Mars-X Express microwave reaction system (CEM, Matthews, NC, USA). The supernatants were cleaned up using Strata SAX cartridges. Then KI was added to the eluent to reduce As^V to As^{III}, which was quantified by HG-AFS using a Millennium Excalibur spectrometer (P S Analytical, Kent, UK). Triplicate runs were carried out and quantification was based on peak height. Validation was carried out using standard reference materials NIST 1568b and ERM BC211 rice flour.

2.3. Outlier identification

Outliers in raw, dry weight (dw)-based iAs data were identified by the John Tukey's method using the following fences:

$$(\mathbf{Q}_1 - 1.5\mathbf{IQR}), \quad (\mathbf{Q}_3 + 1.5\mathbf{IQR}) \tag{1}$$

where Q1 and Q3 are the first and third quantiles of the data set in each crop category, and IQR is the interquantile range:

$$\mathbf{IQR} = \mathbf{Q}_3 - \mathbf{Q}_1 \tag{2}$$

The outliers were excluded before further statistical treatment.

2.4. Health risk assessment

Risk assessment consists of four steps: hazard identification, dose response assessment, exposure assessment, and risk characterization. Non-carcinogenic effects are beyond the scope of this discussion.

The objective of Monte Carlo simulation was to estimate the probability a person develops cancer in lifetime in 19 SSA countries that consumed RTP for >20% of dietary calories. Table 3 lists the population and per-capita annual RTP consumptions of these countries. First, the probability ($p_{c,i}$) a person to reside in a specific country origin i was calculated as follows:

$$\mathbf{p}_{\mathbf{c},\mathbf{i}} = \frac{\mathbf{pop_i}}{\sum_1^{19}\mathbf{pop_i}} \tag{3}$$

where pop_i is the population of SSA country i. The relative contribution of the annual RTP consumption from country i was:

$$\mathbf{Y}_{\mathbf{c},\mathbf{i}} = \mathbf{p}_{\mathbf{c},\mathbf{i}} \times \mathbf{RTP_i} \tag{4}$$

where RTP_i is the mean annual RTP consumption from SSA country i listed in Table 3. The total annual consumption of a person in SSA countries, Y_c, is:

$$\mathbf{Y_c} = \sum_1^{19} \mathbf{Y}_{\mathbf{c},\mathbf{i}} \tag{5}$$

Target lifetime cancer risk, TR, was estimated based on the US. Environmental Protection Agency (EPA) guidance (Lin & Liao, 2008; Liu, Huang, & Hsueh, 2005; Tsuji, Yost, Barraj, Scrafford, & Mink, 2007) as follows:

$$\mathbf{TR} = \frac{\mathbf{Y_c} \times \mathbf{Ed_{tot}} \times \mathbf{MCS_{iAs}} \times \mathbf{CPS_0}}{\mathbf{BW_a} \times \mathbf{AT_c}} \tag{6}$$

where Ed_{tot} is the exposure duration (30 years), MCS_{iAs} (μg g^{-1}) is the concentration of iAs in RTP, CPS_0 is the oral carcinogenic