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Dietary exposure to inorganic arsenic of the Hong Kong population: Results of the first Hong Kong Total Diet Study

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ABSTRACT

Inorganic arsenic, a human carcinogen, can be found in the environment and food. In the first Hong Kong Total Diet Study, the dietary exposure of the Hong Kong people, including various age-gender subgroups, to inorganic arsenic was estimated for assessing the associated health risk. Food samples, which represented the Hong Kong people's diet, were collected and prepared "as consumed" for analysis. Concentrations of inorganic arsenic, as sum of arsenite (As(III)) and arsenate (As(V)) were determined in 600 composite samples by using inductively coupled plasma mass spectrometry. The dietary exposures were estimated by combining the analytical results with the local food consumption data of the adult population. The mean and 95th percentile of inorganic arsenic exposures of the Hong Kong people were 0.22 and 0.38 μg/kg body weight (bw)/day, respectively. Among the 12 age-gender subgroups, the respective exposures ranged from 0.19 to 0.26 μg/kg bw/day and from 0.33 to 0.46 μg/kg bw/day. The main food category that contributed inorganic arsenic was "cereals and their products" (53.5% of the total exposure), particularly rice. Having considered the carcinogenic risk of inorganic arsenic to humans, it is suggested that efforts should be made to reduce the inorganic arsenic exposure of the Hong Kong population.

1. Introduction

Inorganic arsenic is the more toxic form of arsenic, a metalloid, which is found in the environment from both natural occurrence and human activities (JECFA, 2011). Food is recognised as a major source of inorganic arsenic exposure (EFSA, 2009; JECFA, 2011; WHO, 2003). The inorganic arsenic levels found in foods and beverages usually do not exceed 100 μg/kg with mean value of less than 30 μg/kg. However, some food commodities, such as seaweed, rice, some fish and seafood commodities, may contain higher inorganic arsenic levels. The inorganic arsenic as a proportion of the total arsenic also varies among food commodities (Codex, 2011a; JECFA, 2011).

Long term exposure to inorganic arsenic may cause bladder, lung and skin cancers, skin lesions, cardiovascular disease, neurotoxicity, and diabetes (EFSA, 2009; IARC, 2009; JECFA, 2011). In 2010, the Joint FAO/WHO Expert Committee on Food Additives (JECFA) determined the inorganic arsenic Benchmark Dose Lower Confidence Limit for a 0.5% increased incidence of lung cancer in human ($BMDL_{0.5}$) as 3.0 μg/kg body weight (bw)/day (in the region of 2–7 μg/kg bw/day). This $BMDL_{0.5}$ contains the following uncertainties, namely, the assumptions on total exposure extrapolated from the drinking water; and the extrapolation of the $BMDL_{0.5}$ to other populations due to the influence of nutritional status such as low protein intake, and other lifestyle factors on the effects observed in the studied population (JECFA, 2011). $BMDL_{0.5}$ cannot be regarded as a safety reference value. A dietary exposure below the $BMDL_{0.5}$ does not mean that there is no health risk (Codex, 2011b). Therefore, in order to assess the health risk of dietary exposure the margin of exposure (MOE) approach is used.

The MOE for inorganic arsenic is the ratio of $BMDL_{0.5}$ to the inorganic arsenic dietary exposure estimate; the higher the MOE, the lower the health concern, and vice versa. The MOEs can be used for priority setting for risk management actions, and the level of regulatory or non-regulatory intervention can be determined taking account of the size of the MOE (Codex, 2011b).

The Centre for Food Safety (CFS), being the government food safety control authority of Hong Kong, is responsible for assessing the health risks associated with food sold in Hong Kong. The CFS started to conduct its first Hong Kong Total Diet Study (TDS) in 2010 aiming to provide dietary exposure estimates of contaminants and nutrients for the Hong Kong people and various age-gender subgroups. TDS approach was adopted because it has been recognised internationally as the most cost effective way to estimate the dietary exposures to chemicals or nutrients (WHO, 2006). It focuses on substances in the whole diet, not individual foods, and aims to assess dietary exposure to substances actually

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ingested by the population, rather than the substance concentration in food (WHO, 2006).

This article presents the first Hong Kong TDS results of the dietary exposure of people to inorganic arsenic and assessment of its associated health risk. In this study the actual inorganic arsenic levels in food as consumed were determined, rather than using generalised conversion factors to estimate the inorganic arsenic content from total arsenic measurement, which has been applied in some exposure assessment studies (JECFA, 2011).

2. Material and methods

2.1. Food Consumption data and selection of food samples

The food consumption data were taken from the Hong Kong Population-based Food Consumption Survey (FCS) conducted by the CFS in 2005–2007 (FEHD, 2010). Through a quota sampling by gender and age groups, 5008 Hong Kong adults aged 20–84 were invited to complete two non-consecutive 24-h dietary intake questionnaires. The survey results revealed that over 1400 food items were being consumed by the Hong Kong people. When estimating the dietary exposure, a weighting based on age-gender group was applied to adjust for bias arising from the age-gender quotas. The weighting was based on the population distribution by age and gender in the 2006 Population By-census (FEHD, 2011, 2010).

One hundred and fifty most commonly consumed food items were selected for the study, based on the food consumption pattern of the Hong Kong people. In order to cover the whole diet of the Hong Kong people, a food mapping process of the 150 TDS food items over the 1400 FCS food items was carried, and it is described under Section 2.4.

2.2. Food sampling and preparation

To provide greater flexibility in calculating dietary exposure values for various segments of the population, an individual food approach was used in the study (FAO/WHO, 2009). Three samples of each TDS food item were collected and prepared in four occasions from March 2010 and February 2011. They were homogenised individually and combined into a composite sample. A total of 1800 samples were collected and combined into 600 composite samples.

Based on the buying habits of the majority of the Hong Kong people, the samples were collected from a range of retail outlets, such as supermarkets, wet markets, groceries and restaurants, etc., in different areas of Hong Kong. No verification of species/varieties and the country of origin of the samples were made because it was not the focus of the study.

The collected food samples were prepared as food normally consumed, i.e. table ready, in a manner consistent with cultural habits, and the preparation methods ranged from simple rinsing to cooking. Distilled water was used for food preparation. No salt and cooking oil were added during food preparation because salt and cooking oil were also considered as food items selected for inorganic arsenic testing, and the consumption amount had been captured separately (FEHD, 2011). This approach is consistent with the TDS conducted by other countries, such as Australia and New Zealand (FSANZ, 2011; NZFSA, 2011).

Prepared samples were kept at −18 °C and transferred to a laboratory for analysis (normally less than 3 h). The equipment used for preparing and homogenising the composite samples was thoroughly washed between each preparation (e.g. cleaning with a laboratory-grade detergent, rinsing thoroughly with hot tap water, and rinsing thoroughly with deionised water) to avoid the risk of cross-contamination.

2.3. Chemical analysis of inorganic arsenic

Inorganic arsenic refers to the sum of arsenite (As(III)) and arsenate (As(V)). The analytical method used was described by Munoz et al. (1999). In brief, the composite samples (2.5 g) were solubilised in around 3 mL deionised water and 18.4 mL concentrated hydrochloric acid. After reduction with 2 mL hydrogen bromide and 1 mL hydrazine sulphate at 15 mg/mL, arsenite was extracted into 10 mL chloroform twice. The arsenite in the combined chloroform was then back extracted to 10 mL diluted hydrochloric acid. Subsequently, the organic matters remaining in the back-extracted acid solution were removed and destructed by dry-ashing at 425 °C for 12 h. The resulting ash was dissolved in a solution of 0.5 mL deionised water and 5 mL concentrated hydrochloric acid. For quality control, a reference sample was purchased from the Food Analysis Performance Assessment Scheme (FAPAS, York, UK).

The analytical quantification of inorganic arsenic was performed, using a hydride generation inductively coupled plasma mass spectrometry (Agilent ICP-MS 7500-Ce). [Note: This procedure was also known to extract a small amount of monomethylarsenic species (FSA, 2009a).] For the comparison of measured and referenced concentrations of inorganic arsenic, each test run included spiked test samples and a quality control reference material. The limits of detection (LOD) and the limits of quantification (LOQ), respectively, were 3 and 10 μg/kg in food, and 1.5 and 5 μg/kg in drinking water and bottled water.

Recovery percentages (n = 218) based on a quality control reference material (FAPAS T0792 with assigned value of 67.2 mg/kg) ranged between 89 and 122. Correction for recovery percentages was not performed. The average spiking recovery percentage was 87% (RSD of ±7%).

2.4. Dietary exposure estimates

The result reveals that 49% of the test samples were below the LOD. The mean levels of the four occasions for exposure estimation were calculated under the assumption that the non-detectable results equal to the half of LOD, according to the World Health Organization recommendations (WHO, 1995).

The 150 TDS food items were mapped with 1400 food items captured by FCS in order to cover the whole diet of the Hong Kong people. The mean levels of the TDS food items were assigned to the mapped FCS food items with an application of conversion factors taking reference to the differences in water content (FEHD, 2011). To cite an example, cooked white rice in TDS food was mapped to cooked white rice and congee in FCS. As a result, over 99% of the food intake of the Hong Kong people was covered in the dietary exposure estimation after food mapping.

Dietary exposure to inorganic arsenic of individual respondent was estimated by combining the food consumption data with inorganic arsenic level found in mapped food items according to the following formula:

$$E_r = \sum_{i=1}^{n} \left(\frac{F_{r,i} \times C_i}{1000 \times bw_r} \right) \quad (1)$$

where E_r is the total daily dietary exposure to inorganic arsenic of the respondent r (μg/kg bw/day), $F_{r,i}$ is the daily intake of the mapped FCS food item i by the respondent r (g/day), C_i is the inorganic arsenic level of the mapped FCS food item assigned (μg/kg), bw_r is the body weight of the respondent r (kg), n is the total number of mapped FCS food items consumed by the respondent r. The mean exposure level among the respondents after weighting by age-gender is used to represent the average dietary exposures of the Hong Kong people. The 95th percentile exposure level is used to represent the dietary exposure of the high consumer of the Hong Kong people. Dietary exposure estimation was performed with the aid of an in-house developed web-based computer system, EASY (Exposure Assessment System), that takes food mapping and weighting of data into consideration.

3. Results and discussion

The inorganic arsenic contents of 150 TDS food items are given in Table 1. The highest levels were found in water spinach (mean: 74 μg/kg), salted eggs (mean: 58 μg/kg) and oyster (mean: 58 μg/kg). The high level of inorganic arsenic in water spinach may be due to the aquatic growing conditions. The presence of inorganic arsenic in salted eggs may be due to the use of plant ash and/or loess (a kind of light-coloured soil) in the production process, and its presence in oyster may be due to the environmental contamination. On the other hand, all samples of "dairy products" and "fats and oils" were not detected with inorganic arsenic, and 95% of non-alcoholic beverage samples including water samples were also not detected with inorganic arsenic. Although it has been reported that water is one of the most significant sources of inorganic arsenic exposure (JECFA, 2011; WHO, 2003), water samples from this study, including drinking water and bottled water collected at various parts of Hong Kong, were not detected with inorganic arsenic.

Dietary exposure to inorganic arsenic was estimated by using the inorganic arsenic level found in food items together with the food consumption data. The estimated mean inorganic arsenic dietary exposure of the Hong Kong people was 0.22 μg/kg bw/day and its 95th percentile was 0.38 μg/kg bw/day. For adult males and females (aged 20–84), the mean inorganic arsenic dietary exposure values were 0.23 and 0.21 μg/kg bw/day respectively. Among the 12 age-gender groups, the mean inorganic arsenic dietary exposures of the individual subgroups ranged from 0.19 μg/kg bw/day (female aged 20–29) to 0.26 μg/kg bw/day (male aged 60–69) and the 95th percentile range from 0.33 μg/kg bw/day (female aged 20–29) to 0.46 μg/kg bw/day (male aged 60–69) (Fig. 1). For illustration, using the average body weight of the Hong Kong people (estimated to be 61 kg), a 61-kg person is estimated to be