



A comprehensive assessment of arsenic in commonly consumed foodstuffs to evaluate the potential health risk in Bangladesh

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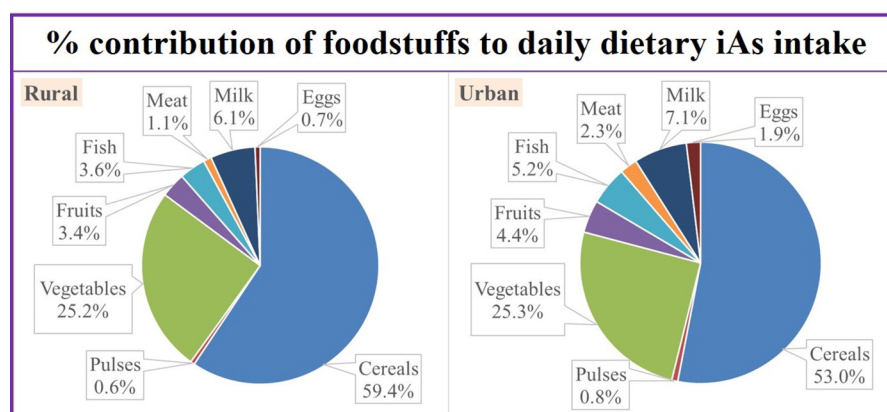
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HIGHLIGHTS

- A comprehensive health risk assessment from dietary arsenic exposure was evaluated.
- Sample collected from 30 agro-ecological zones for the first time in Bangladesh.
- Rural and urban adults are consuming more arsenic through food than the safe limit.
- Cereals, vegetables, milk, and fish contribute about 90% to EDI of inorganic As.
- Inhabitants are exposed chronically to arsenic induced risks.

GRAPHICAL ABSTRACT



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ABSTRACT

Arsenic (As), particularly of its inorganic form (iAs) is highly toxic, and its presence in food composites is a matter of concern for the public health safety, specifically in Bangladesh which is regarded as the most arsenic affected country throughout the world. This study was carried out to investigate the levels of As in the composite samples of commonly consumed foodstuffs collected from 30 different agro-ecological zones for the first time in Bangladesh. Most of the individual food composites contain a considerable amount of As which was, as a whole, in the range of 0.077–1.5 mg/kg fw which was lower than those reported from Spain, EU, France, Korea, whereas higher than those of Mexico, Chile, Japan, Cambodia, Hong Kong, Serbia, respectively. Cereals, vegetables, milk, and fish contribute about 90% to the daily intake of inorganic arsenic. Human health risk of dietary iAs was assessed separately for both the rural and urban adults. The estimated daily dietary intakes (EDI) of iAs for the exposed rural (3.5) and urban residents (3.2 µg/kg-BW/day) clearly exceeded the previous provisional tolerable daily intake (PTDI) value of 2.1 µg/kg-BW/day, recommended by the World Health Organization (WHO). From the health point of view, this study concluded that both the rural and urban residents of

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1. Introduction

Arsenic (As) is omnipresent in almost each and every compartment of the environment that can pollute soil, water, and biota and ultimately affects human health and well-being (Bundschuh et al., 2012; Culioli et al., 2009). Arsenic contamination in the ground water has been reported worldwide (Nordstrom et al., 2002). The contaminated ground water with higher levels of arsenic compared to surface water is being mainly used as drinking water and irrigation for agriculture and aquaculture practices (Bhattacharya et al., 2010; USEPA, 1989). Arsenic can be taken up by biota, which is either consumed by humans or incorporated into fodder for poultry and livestock (Bundschuh et al., 2012). Although the groundwater contamination by arsenic is already considered as a serious and severe global environmental problem, but little is known about diet as an additional source of arsenic exposure. Recently, diet studies that included evaluations of total arsenic (tAs) and inorganic arsenic (iAs) in foods have been reported for many countries (Diaz et al., 2015; Wang et al., 2013; Wong et al., 2013; Arain et al., 2009; Li et al., 2006; Muñoz et al., 2005; Del Razo et al., 2002). The total dietary study performed by US Food and Drug Administration (FDA) indicated that food contributes 93% of the total intake of arsenic (Adams et al., 1994).

Arsenic in which humans are exposed can be both inorganic and organic forms. Chronic exposure to inorganic arsenic creates several disorders on different biological systems including digestive, respiratory, cardiovascular, hematopoietic, endocrine, renal, neurological, and reproductive, which ultimately lead to cancer (Santra et al., 2013; Maharjan et al., 2005). Daily intake of 1 mg inorganic arsenic may give rise to skin effects within a few years (WHO, 1981). Drinking 1 L of arsenic contaminated water (50 g/L) per day can create life risk from cancer of 13 per 1000 persons (Smith et al., 1992). The U.S. National Research Council concluded that exposure of 50 g/L of arsenic to human could easily result in a combined cancer risk of 1 in 100 persons (NRC, 1999). The various effects of arsenic poisoning on human health are melanosis, leuco-melanosis, keratosis, hyperkeratosis, dorsum, non-petting oedema, gangrene and skin cancer. Some of these human health effects are currently being observed in populations in South and South-eastern Asia, particularly in countries such as Bangladesh, India and Taiwan (Islam et al., 2014; Halder et al., 2014).

Arsenic contamination is one of the most important issues regarding public health and safety in Bangladesh which is regarded as the most arsenic affected country in the world. According to Hopenhayn (2006), arsenic is probably the contaminant that induces the highest risk of morbidity and mortality worldwide, due to its toxicity level and the number of exposed people. Groundwater is the main source of water for drinking, cooking and domestic use for Bangladeshis. About 27 million Bangladeshis drink water with greater than 50 $\mu\text{g L}^{-1}$ arsenic and 50 million people drink water with greater than 10 $\mu\text{g L}^{-1}$ arsenic (Smith et al., 2000). Moreover, groundwater is continuously being used for irrigation to cultivate a variety of crops, vegetables and for fish culture, including hatchery operation in Bangladesh. Irrigation with arsenic-enriched groundwater is one of the main pathways for arsenic to enter the human food chain (Das et al., 2004; Meharg and Rahman, 2003; Alam et al., 2003) and this has led to a number of studies on transfer of arsenic through the water-soil-crop-food system (Fig. S1). For example, Das et al. (2004) reported arsenic accumulation in agricultural products cultivated using arsenic-enriched groundwater. Some studies showed that fish make the largest contribution of arsenic to the diet. Kar et al. (2011) stated the health risks from human intake of aquaculture fish with arsenic contamination. It was observed that

arsenic was bioaccumulated in fish from water (Larocque, 1998), whereas arsenic accumulation in fish is higher than in water (Arain et al., 2008; Liao and Ling, 2003).

The studies of arsenic in human food chain have been done occasionally from specific areas or food systems in Bangladesh using different methodologies (Islam et al., 2014; Saha and Zaman, 2013; Smith et al., 2006; Williams et al., 2006; Alam et al., 2003). Hence, they are not necessarily comparable and results are inconclusive. To estimate the public health risk, it is necessary to analyze arsenic at different food chains of the whole country within the same period and using the same methodology. It is well known that the levels of arsenic in foods are generally accumulated from the local environments, indicating the foodstuffs grown in arsenic-enriched rural areas are contaminated with arsenic in Bangladesh. In fact, most of the foodstuffs available in urban markets and commonly consumed by the urban residents originated from the rural areas. Hence, the identifications of daily dietary intake of arsenic and associated health risk assessment are urgent for both the rural and urban residents in Bangladesh.

This study assessed arsenic levels in food composites covering all the agro-ecological zones of Bangladesh and daily dietary intake for both of its rural and urban residents. The potential health risk was assessed through dietary intakes of commonly consumed food and drinking water. To our knowledge, this is the first study to investigate the dietary intake of arsenic and associated health risk at the population level (considering 30 agro-ecological zones of Bangladesh) based on the consumption of daily common foodstuffs by the Bangladeshi adults.

2. Materials and methods

2.1. Sample collection and preparation

The per capita per day intake of major food items and intakes of individual food items (BBS, 2011) used to characterize the food consumption pattern of both the rural and urban population in Bangladesh. Both findings showed almost same food composition but different consumption pattern for rural and urban residents. Fresh samples of cereals (*Oryza sativa*, *Triticum aestivum*), pulses (*Lens culinaris*, *Cicer arietinum*), vegetables (*Solanum melongena*, *Daucus carota*, *Phaseolus vulgaris*, *Solanum tuberosum*, *Solanum lycopersicum*, *Allium cepa*, *Capsicum annum* L.), fruits (*Musa paradisiaca*, *Mangifera indica*, *Artocarpus heterophyllus*), freshwater fish (*Labeo rohita*, *Pangasius pangasius*, *Oreochromis mossambicus*), meat (beef, mutton, chicken, duck), egg (chicken, duck), milk (cow) were collected from different rural and urban markets occupying the 30 agro-ecological zones (Fig. 1 and Table S1) of Bangladesh in 2013. These foodstuffs are highly consumed by the Bangladeshi population as their daily diet (BBS, 2011). A composite of at least 10 samples for each food item was prepared and homogenized in a food processor and 50 g test portions were stored at $-20\text{ }^{\circ}\text{C}$ in the Laboratory of the Institute of Nutrition and Food Science (INFS), University of Dhaka. The pre-processed samples were then brought to the Laboratory of Environment and Information Sciences, Yokohama National University, Japan and analyzed for arsenic content, expressed as mg/kg fresh weight (wt.).

2.2. Analytical methods

2.2.1. Reagents and sample extraction procedure

All solutions were prepared with analytical reagent-grade chemicals and ultrapure water. An arsenic (As) standard stock solutions (100 mg L^{-1}) for the determination of the total arsenic (tAs) was

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