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Arsenic species in wheat, raw and cooked rice: Exposure and associated health implications

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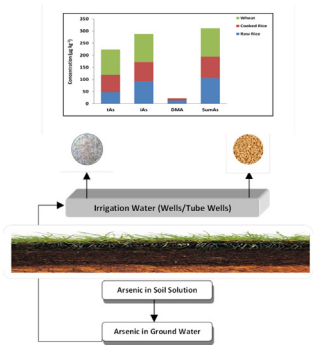
HIGHLIGHTS

- Chronic non-cancer health risks among 97% of study participants due to inorganic arsenic intake from wheat.
- Wheat grown in arsenic affected area poses higher risk than rice as a exposure source.
- Total daily intake of inorganic arsenic above the limit of 2.1 μg kg^{-1} day^{-1} body weight in 74% of participants.
- Above 95% of the children at significantly higher risk due to inorganic arsenic exposure from cooked rice.

GRAPHICAL ABSTRACT



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ABSTRACT

Arsenic concentrations above 10 μg L^{-1} were previously found in 89% of ground water sources in six villages of Pakistan. The present study has ascertained the health risks associated with exposure to total arsenic (tAs) and its species in most frequently consumed foods. Inorganic arsenic (iAs) concentrations were found to be 92.5 ± 41.88 μg kg^{-1}, 79.21 ± 76.42 μg kg^{-1}, and 116.38 ± 51.38 μg kg^{-1} for raw rice, cooked rice and wheat respectively. The mean tAs concentrations were 47.47 ± 30.72 μg kg^{-1}, 71.65 ± 74.7 μg kg^{-1}, 105 ± 61.47 μg kg^{-1}. Wheat is therefore demonstrated to be a significant source of arsenic exposure. Dimethylarsinic acid was the main organic species detected in rice, whilst monomethylarsonic acid was only found at trace levels. Total daily intake of iAs exceeded the provisional tolerable daily intake of 2.1 μg kg^{-1} day^{-1} body weight in 74% of study participants due to concurrent intake from water (94%), wheat (5%) and raw rice (1%). A significant association between tAs in cooked rice and cooking water resulted in tAs intake 43% higher in cooked rice compared to raw rice. The study suggests that arsenic intake from food, particularly from wheat consumption, holds particular significance where iAs is relatively low in water. Chronic health risks were found to be significantly higher from wheat intake than rice, whilst the risk in terms of acute effects was below the USEPA's limit of 1.0. Children were at significantly higher health risk than adults due to iAs exposure from rice and/or wheat. The dietary exposure of participants to tAs was attributable to staple food intake with ground water iAs <10 μg L^{-1}, however the preliminary advisory level (200 μg kg^{-1}) was achievable with rice consumption of ≤200 g day^{-1} and compliance with ≤10 μg L^{-1} iAs in drinking water. Although the daily iAs intake from food was lower than total water intake, the potential health risk from exposure to arsenic and its species still exists and requires exposure control measures.

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1. Introduction

Arsenic (As), a naturally occurring metalloid, is widely present as an environmental contaminant and enters the food chain mainly from contaminated water (European Food Safety Agency, 2009) and several widely consumed foodstuffs (Feldmann and Krupp, 2011; Jiang et al., 2015). Seafood has been identified as the main source of organic arsenic (e.g., arsenobetaine and arsenosugars) and is believed to be non-toxic (Taylor et al., 2017; International Agency for Research on Cancer, 2012b). Most exposure and toxicological assessments have focused on inorganic arsenic (iAs) in drinking water. It is yet not fully understood whether exposure to arsenic via most frequently consumed food (e.g. rice and wheat) causes the same health implications as exposure through drinking water.

Exposure from rice has been assessed in a number of studies (U.S. Food and Drug Administration, 2016; Sand et al., 2015; Chen et al., 2016; Davis et al., 2017; Sun et al., 2012). These studies indicate that rice is the most common exposure source for food stuffs. Rice crops have a comparatively higher tendency to take up iAs as they are grown in submerged soil conditions. Among populations not exposed to iAs via drinking water, rice contributes significantly to the iAs intake (Davis et al., 2017).

Wheat is also an important staple food with a worldwide consumption of 730.9 million tonnes, greater than the 506.5 million tonnes of rice consumed annually (Food and Agriculture Organization, 2017). Past studies have reported lower arsenic levels in wheat than rice (Williams et al., 2007b; Su et al., 2010; Bhattacharya et al., 2010) and provided an impetus to further investigate the health risks due to consumption of wheat grown in arsenic affected regions.

Inorganic arsenic is a recognized carcinogen and its chronic exposure has been reported to result in increased risk of bladder, lung, and skin cancer, type 2 diabetes, and cardiovascular disease (International Agency for Research on Cancer, 2012b).

Organic arsenic compounds are considered less toxic than iAs but should still be included in exposure assessments. Since toxicity depends on the chemical forms of arsenic, arsenic speciation in rice and wheat can provide useful information for risk assessment and management. The Joint Food and Agriculture Organization and the World Health Organization (FAO/WHO) Expert Committee on Food Additives has set, in 2014, advisory levels of 200 μg kg^{-1} iAs in polished rice grains (Codex Alimentarius Commission, 2014). Apart from the EU regulations (EU) 2015/1006) (European Commission, 2015) on adopting this limit, several countries have still not implemented this limit and are in the process of setting regulatory limits for rice based products. Adoption of this advisory limit in different geographical regions requires exposure assessment via rice. Considering these facts, this study has determined the concentrations of total arsenic (tAs) and As species in wheat, raw and cooked rice to assess the relative contribution of dietary arsenic to aggregate daily exposure. Human health hazards associated with daily consumption of rice, wheat and household groundwater by children (age ≤ 16 years) and adults (age > 16 years) was calculated based on these exposures to provide an indication of hazard of each exposure source.

2. Materials and methods

2.1. Study area and study participants

The study villages were located within four districts of Pakistan (Kasur, Sahiwal, Bahawalpur and Rahim Yar Khan), where arsenic concentrations above 10 μg L^{-1} were previously found in 89% of household ground water sources. The sampling frame consisted of 223 households comprising 398 volunteers enrolled and interviewed in our previous studies aimed to assess household ground water arsenic concentrations (Rasheed et al., 2017a) and dietary consumption patterns (Rasheed et al., 2017b). Thus, data on age (3–80 years, mean 36 ± 17 years), gender (246 men and 149 women), body weight (56.6 ± 19.9 Kg), occupation (n = 186 farmers and agriculture labour), cooked rice (469 ± 202 g day^{-1} $person^{-1}$) and wheat intake (372 ± 119 g day^{-1} $person^{-1}$) were obtained by questionnaire from 398 participants in the 223 households enrolled in our earlier study (Rasheed et al., 2017b). The households ground water sources (n = 228) used both for the drinking and food preparation were found to have geometric mean (GM) iAs concentration as 55.33 μg L^{-1} (range: 0.48–3090 μg L^{-1}) and associated daily total water intake of 15.4 μg day^{-1} (0.02–262.57 μg day^{-1}) (Rasheed et al., 2017a).

Wheat and rice was sampled from the households. Raw rice samples were collected from 105 households of villages (Chak-46/12-L, Chak-48/12-I and Chak 49/12-l, Badarpur, Basti Balochan and Kotla Arab), while cooked rice samples could be obtained from 24 households. Twelve households provided paired rice samples (both raw and cooked). The main occupation in the study villages was wheat farming with 47% of 398 study participants engaged in this work (Rasheed et al., 2017b), thus, wheat consumed in the villages was cultivated locally. Following the sampling strategy described by Cubadda et al. (2010), wheat grain samples (n = 189) from two of the most cultivated wheat varieties were collected from the households of six villages. Individual samples (150 g each) were pooled into 8 composite samples weighing in the range of 0.9–7.5 kg.

2.2. Samples collection procedure

For raw rice and wheat samples, sterile re-sealable airtight polyethylene zip lock bags were used, whereas for cooked rice (100 g) 2 oz. polyethylene sterile containers were used. After collection, raw rice (250 g) and wheat samples (150 g) were stored at room temperature, while cooked rice samples were kept in an insulated cooler containing ice in the field and later stored at −20 °C. Cooked rice samples were shipped to Brooks Applied laboratory, USA by FedEx courier with dry ice under strict quarantine regulations and stored at −20 °C prior to analyses. Raw rice and wheat samples were shipped and stored at ambient temperature (20 °C) until analysis in National water quality laboratory Pakistan and Brooks Applied laboratory (BAL), USA.

2.3. Treatment of rice and wheat samples for total arsenic

Rice and wheat samples were rinsed with deionized water (DIW) to remove dust and then dried by air flow at room temperature. Dried samples were milled to powder in a pre-cleaned commercial blender with stainless steel blades. Following USEPA method 3050b (United States Environmental Protection Agency, 1996), a representative 1–2 g (wet weight) or 1 g (dry weight) sample was digested with repeated additions of nitric acid (HNO_3) and hydrogen peroxide (H_2O_2). The resultant digest was reduced in volume while heating at 95 °C ± 5 °C and then diluted with ultrapure water to a final volume of 100 ml and subjected to analysis.

2.4. Treatment of rice and wheat samples for arsenic speciation

Microwave-assisted HNO_3 digestion for arsenic speciation involved adding 0.35 g of ground raw or cooked rice and wheat samples separately into 15 ml sample tubes. 10 ml of 0.16 M suprapure HNO_3 was added to the tube and left to stand overnight. Microwave irradiation was performed with the temperature profile as: 3 min ramp to 55 °C, 10 min at 55 °C, 2 min ramp to 75 °C, 10 min at 75 °C, 2 min ramp to 95 °C, 30 min at 95 °C. The extracts were centrifuged (10 min, 8000 rpm, 4 °C) and the supernatants filtered through a 0.22 μm filter. The filtrate was stored at 4 °C and analyzed within 24 h to minimize any species inter-conversion. For final analysis, 0.1 ml of the filtered solution was combined with 0.9 ml of DIW in a 1.5 ml vial and mixed for 10 s with a vortex mixer (D'Amato et al., 2011; Raab et al., 2009b).

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