Studies on chronic exposure to arsenic toxicity through food chain and drinking water with respect to rice grain contamination in selected sites of West Bengal and search for mitigation options

> Synopsis submitted by MADHURIMA JOARDAR Index No.: D-7/ISLM/09/18

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2022

Presence of arsenic (As), a naturally occurring toxic metalloid (human carcinogen) in groundwater has been a recurring problem worldwide. Arsenic contamination along with its consumption poses considerable health exposure and risk to differently exposed populations. Perception of health risk has been assessed on inhabitants being exposed to As through dietary foodstuffs (drinking water, rice and vegetables). One of the most crucial environmental concerns is natural groundwater As contamination in rural Bengal and its resulting toxic effect poses threats to human health. Foremost, the study highlights the As contamination scenario in the As-affected Gaighata, Deganga and Swarupnagar blocks of North 24 Parganas district, West Bengal, India. The mean As concentration of domestic shallow tube-well water is 5.25 and 28.7 fold higher than the mean As of deep tube-well water from Gaighata and Deganga blocks, respectively. About 78.5 and 70% of the deep tube-well groundwater samples from Gaighata (depth: 177-238 m) and Deganga (depth: 152-244 m) blocks, respectively, have been identified with As concentration less than 10 µg/L, the recommended WHO value. The groundwater quality index (WQI value) showed that 36% and 25% of deep tube-well water samples are characterized under 'good water quality', recommended for drinking purpose in the respective Gaighata and Deganga block. In a similar way, Swarupnagar block, covering 9gram panchayats showed both As and iron (Fe) categorized under considerable contamination scenario, whereas, quantitative estimation of ecological risk has been categorized under very high risk class. Likewise, Gaighata block, comprising of 13-gram panchayats showed considerable As and high Fe contamination of the deep tube-well water sources, along with high ecological risk factor.

Apart from As-contaminated drinking water, rice As contamination and its consumption poses a significant health threat to humans. Rice grain which is considered as a staple food crop of rural Bengal, poses an additional danger to differently exposed inhabitants. Health risk through post-monsoonal rice cultivars is lower compared to pre-monsoonal rice cultivars. Moreover, the health risks through unpolished rice are higher compared to the polished rice cultivars. The study also focuses on the contribution of As, micronutrients (zinc and selenium) and its associated benefit-risk assessment through cooked rice from rural (exposed and control area) and urban (apparently control area) populations. The mean decreased percentages of As from uncooked to cooked rice for exposed (Gaighata), apparently control (Kolkata), and control (Pingla) areas, are 73.8, 78.5, and 61.3%, respectively. The margin of exposure through cooked rice (MoE cooked rice) < 1 signifies the existence of health risk for all the studied exposed and control age groups. Intake of inorganic As (iAs) is measured as a probable area of concern

depending on health risk. Rice contributes approx. 89.5% of iAs, which is being nurtured in exposed areas and transported to control areas. The respective contribution of iAs in uncooked and cooked rice are nearly 96.6, 94.7, 100% and 92.2, 90.2, 94.2% from exposed, apparently control and control areas. LCR analysis for the exposed, apparently control, control population (adult male: 2.1x10⁻³, 2.8x10⁻⁴, 4.7x10⁻⁴; adult female: 1.9x10⁻³, 2.1x10⁻⁴, 4.4x10⁻⁴; and children: 5.8x10⁻⁴, 4.9x10⁻⁵, 1.1x10⁻⁴) through cooked rice is higher than the recommended value i.e. 1×10^{-6} , respectively. Whereas, HQ>1 has been observed for all age groups from the exposed area and adult male group from the control area. Adults and children from rural area showed that ingestion rate (IR) and concentration are the respective influencing factors towards cooked rice As, whereas, IR is solely responsible for all age groups from urban area. A vital suggestion is to reduce the IR of cooked rice for control population, to avoid the As-induced health risks. The average intake ($\mu g/day$) of micronutrients is in the order of Zn>Se for all the studied populations and Se intake is lower for the exposed population (53.9) compared to the apparently control (140) and control (208) population. Benefit-risk evaluation supported that the Se-rich values in cooked rice are effective in avoiding the toxic effect and potential risk from the associated metal (As).

On a subjective note, the biomarkers of the As (urine, scalp hair and nail) were analyzed to put forward the effectiveness of acute and chronic As exposure and its level of toxicity on human health. Health exposure and perception of risk assessment have been evaluated on the populations exposed to different As levels in drinking water (615, 301, 48, 20 µg/L), rice grain (792, 487, 588, 569 µg/kg) and vegetables (283, 187, 238, 300 µg/kg) from four villages in As endemic Gaighata block, West Bengal. Dietary As intake rates for the studied populations from extremely highly, highly, moderately, and mild As-exposed areas were 56.03, 28.73, 11.30, and 9.13 µg/kg bw/day, respectively. Acute and chronic effects of As toxicity were observed in ascending order from mild to extremely highly exposed populations. Statistical interpretation using 'ANOVA' proves a significant relationship between drinking water and biomarkers, whereas "two-tailed paired t test" justifies that the consumption of As-contaminated dietary intakes is the considerable pathway of health risk exposure. According to the risk thermometer (SAMOE), drinking water belongs to risk class 5 (extremely highly and highly exposed area) and 4 (moderately and mild exposed area) categories, whereas rice grain and vegetables belong to risk class 5 and 4, respectively, for all the differently exposed populations. The carcinogenic (ILCR) and non-carcinogenic risks (HQ) through dietary intakes for adults were much higher than the recommended threshold level, compared to the children. Another research observation also reveals that the different age groups of children are sub-clinically affected as a result of As toxicity through drinking of contaminated water. The rate As intake through drinking water by children of respective age groups as 1-5 years, 6-10 years and 11-15 years has been observed as 3.74, 5.02 and 5.04 μ g/kg bw/day, that is more than the WHO acceptable value of TDI i.e. $3 \mu g/kg bw/day$. The prominent level of As in urine signifies that the studied groups of children are presently exposed to As-contaminated drinking water. Amongst the different age-groups of children, nail As appears to be higher compared to hair As by 1.75 (1-5 years), 1.59 (6-10 years) and 1.58 (11-15 years) times, respectively. Human health risk is dependent on the intake of inorganic As which is measured as potential concern worldwide. The observed mean cancerous risk is high for the respective age groups of children i.e. (1-5 years: 3.9x10⁻⁴), (6-10 years: 1.1×10^{-3}) and (11-15 years: 1.7×10^{-3}) through drinking water. Likewise, the mean noncancerous (HQ value) risk for children of 1-5 years is 0.88, which is lower than the allowable value (HQ>1). Whereas, the HQ for6-10 years and 11-15 years' children are 2.43 and 3.85, respectively, which are much higher than the recommended level. Precisely, investigation has been carried out evaluating acute and chronic As exposure of the school children from highly exposed and apparently control area of Bengal and their future risk of sufferings. This study reveals a potential area of concern related to sub-chronic exposure in children due to As toxicity through contaminated drinking water and rice. The daily dietary intake of As through drinking water, rice, and mid-day meal for the school children from the exposed area is 366 µg which is higher compared to the apparently control area (166 µg). The dietary intake rate of As through drinking water for the exposed children is 5.26 µg/kg bw/day which is much higher compared to the WHO recommended value of TDI i.e. 3.0 µg/kg bw/day. Higher accumulation of As has been observed in the biomarkers (urine, hair, and nail) for the exposed children (64.6 µg/L, 2656 and 4256 µg/kg, respectively) compared to apparently control children (4.35 µg/L, 1286 and 1805 µg/kg, respectively). The cancer risk assessment is high through the consumption of As-contaminated drinking water for the exposed children compared to the apparently control children. Intake of inorganic As is considered as a potential area of concern depending on human health hazard. Contribution of inorganic As is high (appx. 89.5%) through the consumption of contaminated rice grains, which are being cultivated in exposed areas and transported to control areas. The probability of cancer risk is almost equally distributed through ingestion of contaminated rice grains for both the studied populations. An awareness campaign was organized for the exposed school children in the school campus on the As pollution and directed them to consume As-safe water. The pond (surface water) was chemically treated by slow sand filtration process, monitored by adding disinfectant like bleaching powder and supplied to the inhabitants and maintained by a local non-Government organization (Madhusudankati Krishak Kalyan Samity) collaborating with 'Sulabh International Social Service Organization' located in Madhusudankati village has been used by the school authority and children in domestic scale for drinking and cooking activities. A follow-up health exposure study was conducted after 8 months on the exposed school children. The survey highlights a noteworthy declining trend of As accumulation in urine and other biological tissues of the exposed children after minimizing the level of As contamination. Interestingly, chronic arsenicosis patients have been exposed to treated drinking water from the year 2016 supplied through a surface water treatment plant which has been executed in the year 2014 and maintained by a local non-Government organization (Madhusudankati Krishak Kalyan Samity) collaborating with 'Sulabh International Social Service Organization' located in Madhusudankati village. A longitudinal health effect study was carried out on a group of chronic arsenicosis patients (aged 42-75 years including 17 males and 7 females) for one year (2018-2019) to highlight impact of treated (surface water) mainly used for drinking water by the arsenicosis patients along with continuous consumption of contaminated dietary foodstuffs. The daily dietary intake rate of As through cooked rice is 5.50 µg/kg bw/day, which is much higher than TDI recommended value compared to cooked vegetables and treated drinking water. The effect of acute toxicity showed a decreasing trend of 42.9% As in urine (n = 24)after 6 months. Scalp hair (n = 19) and nail (n = 18) As concentration showed a decreasing trend of 39.3% (range: 1.34-86.2%) and 36.9% (range: 0.88-85%), respectively after 12 months. The body hair (hand and leg) and skin scale As accumulation showed high and diverse distribution pattern. Excretion of As through sweat was higher than urine with a mean concentration of 34.7 µg/L (range: 4.76–65 µg/L). Chronic As exposure for a long period of time is the considerable pathway to severe dermatological skin manifestations in the arsenical patients. One-way ANOVA (Tukey-test) interpretation showed a significant relationship between As intakes, biological tissues and dermatological manifestations within the studied groups. Linear mixed modelling showed differential temporal trends of As levels through biomarkers for both studied male and female patients. The SAMOE value for treated drinking water and cooked vegetables showed low to moderate concern level (class 3), whereas, high concern level (class 5) was observed for cooked rice. The future cancer and non-cancerous risk predominantly exist through consumption of rice compared to vegetables and treated drinking water.

As a mitigation strategy, different cooking methodologies has been used to reduce As in cooked rice. Firstly, field-based co-precipitation process showed 53% of As-reduction in cooking water after 24 h. Cooking with water after co-precipitation process i.e. 24h showed 48.6 and 59.1% As reduction in cooked rice using respective ratio of 1:3 and 1:6. Further, 1 h soaking contact time with As-safe water showed 41.9% As reduction in soaked rice followed by cooking with As-safe water reduces 70% of As in cooked rice. An attempt has been taken to resist As in cooked rice using chelating agents like citrate and tartrate solution at the time of soaking using photo-catalysis method. Since, huge availability of As-safe water is of serious concern in As-prone areas, so use of citrate and tartrate solution (1:10) at the time of soaking followed by cooking with As-safe water reduces As up to 50.1% and 48.5%, respectively maintaining the domestic scale cooking ratio of 1:3.

Overall, the study focusses on supply of As-safe water to the As-exposed populations mainly for drinking purposes through proper surface water treatment plant to prevent severe health risks. Use of As-contaminated groundwater must be avoided during agricultural as well as cooking practices to restrict As entry in crops (rice) and vegetables leading to prevention of food chain contamination. Presence of micronutrients in rice is highly effective in avoiding the toxic effect and potential risk through As. So, promoting awareness program on As severity and proper healthy nutritional food is highly recommended for the endangered population to fight against the devastating calamity of As.