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Modeling Dietary Intake of Arsenic and the Associated Human Health Risk for People Living in Rural Bangladesh

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Abstract: Dietary intake of Arsenic (As) via contaminated drinking water and food is the major exposure pathway for people living in rural Bangladesh and causes adverse human health effects where groundwater As contamination and the associated human health risks are potentially spatially connected. This research quantifies the spatial variability of As in the rural landscape in relation to As concentrations in drinking water, rice and other foods. The impact of As exposure on an individual's human health was quantified using the average daily intake (ADI) to calculate the Hazard Quotient (HQ) and Cancer Risk (CR) using Bangladesh specific parameters via integration of a number of complex databases using SQL queries and spatial modeling. The As concentration in tubewell water ranged from 0 to 808 $\mu\text{g L}^{-1}$ with a mean of 48 $\mu\text{g L}^{-1}$ indicating that a large proportion of the population (73%) were exposed daily to $>10 \mu\text{g L}^{-1}$ As via drinking water. Arsenic concentrations were highly spatially variable even for water sources located within close proximity ($< 50 \text{ m}$). A typical adult Bangladeshi diet consisted of a large volume of water ($2.6 \pm 1.4 \text{ L day}^{-1}$), together with a large portion of rice ($1494 \pm 776 \text{ g FW day}^{-1}$), vegetables ($136 \pm 37 \text{ g FW day}^{-1}$) and dal ($35 \pm 2 \text{ g FW day}^{-1}$), supplemented by irregular consumption of small amounts of fish or meat. Dietary pattern analysis indicated that, the rural diet was primarily reliant on water (53%) and cooked rice (31%) supplemented by smaller amounts of vegetables (3%), dal (1%), fish (1.5%), meat (1.5%) and other foods (9%). The ADI of As was $1.98 \mu\text{g kg}^{-1} \text{ BW day}^{-1}$ ($n=345$), and both HQ and CR were significantly higher than acceptable risk levels.

Keywords: Arsenic, dietary intake, average daily intake (ADI), risk, Bangladesh.

1 Introduction

Groundwater tubewells (TW) were introduced to rural communities of Bangladesh by UNICEF in the late 1970s to prevent human water borne diseases. Consequently, today most rural populations obtain 'safe' drinking water from hand-pumped shallow tubewells (STW). However, many of these tubewells were subsequently shown to be contaminated with Arsenic (As) and ingestion of contaminated water was identified as a major As exposure pathway (Chen and Ahsan 2004; Watanabe et al., 2004; Khan, 2009). Since food crops in Bangladesh are often grown using As contaminated irrigation water, food consumption is also an important route for human As exposure (Meharg, et al., 2003; Williams et al., 2006; Khan, 2009) and dietary intake of both contaminated food and water has been identified as the major human As exposure pathway in the rural landscape (Watanabe et al., 2004; Kile et al., 2007; Khan, 2009). Prolonged dietary exposure to high levels of As causes skin lesions, melanosis, hyperkeratosis, jaundice, vascular diseases and skin, lung, and bladder cancers (Yu et al., 2003; Tchounwou et al., 2004; Ahsan et al. 2006). The interactions between dietary intake patterns, demography, socio-economic status and As concentration in various environmental media provides a complete picture of the extent and severity of As exposure in a

given landscape. In this paper these interactions are examined for the rural population of Bangladesh to estimate the dietary intake of arsenic and associated human health risk.

2 Study area and method

2.1 Study area

This study involved sixty households from six villages located in the Sirajdikhan *upazila* of the Munshiganj district. Spatial extent of the Sirajdikhan *upazila* is 23°30' - 23°41' latitude and 90°14' - 90°27' longitude with an area of 180 km² located mainly within the old Brahmaputra floodplain bounded by the Ichamati and Dhaleshwari rivers in the north.

2.2 Collection of dietary food and water intake and other exposure parameters

The village and household selection criteria, together with individual householder's dietary intake of food and water were collected using a food frequency questionnaire (FFQ) previously described (Khan et al., 2009a and 2009b). Concurrent with dietary information, drinking water and food samples were also collected for analysis together with body weights. Dietary information was collected from 345 householders including adult males (n=125), adult females (n=139) and children (n=81).

2.3 Methods for analysis of As in water and food

2.3.1 Total As analysis - water samples

Water samples collected in the field (50 mL) were filtered through disposable 0.45 µm cellulose acetate filters, acidified with nitric acid (3 drops) and refrigerated at 4°C until total As was analysed directly using ICP-MS.

2.3.2 Processing of food / vegetables samples

Foods collected in the field were returned to the laboratory under cool conditions and refrigerated at 4°C until processed. All samples were washed clean of any adhering dirt, cut into small segments and dried to constant weight at 60°C before being finely powdered. Subsamples of dried and ground food (0.5 g) were digested at 140°C with concentrated nitric acid (5 mL) using a temperature controlled digestion block (AI Scientific Block Digestion System AIM 500). Digested samples were diluted with 0.1% HNO₃ (20 mL), mixed thoroughly by vortexing and filtered through Whatman No.42 filter papers prior to analysis for total As by ICP-MS. For quality assurance and control each batch included blanks, duplicates, spikes and certified reference materials (CRMs) at a rate of 5%. The certified reference material, Bush Branches and leaves (GBW07603) had an As content of 1.25 ± 0.15 µg g⁻¹. All As recoveries were > 90 % with a RSD < 5%. Selected plant samples were also spiked with an As standard prior to digestion to give a concentration of 100 ppb (200 µL of 10 ppm standard in 20 mL). Spike recoveries were all > 95% indicating few plant matrix effects. Inorganic As (IAs) was calculated as 80% of the total As (Williams et al., 2006). The mean arsenic concentration of rice and other food items was reported on a fresh weight (FW) basis using moisture contents determined at the time of sampling.

2.4 Estimation of average dietary intake (ADI) of As

Individual average daily intake (ADI) of As from water, rice and other foods was calculated using:

$$ADI = \frac{C \times IR \times EF \times ED \times BAV}{BW \times AT} \quad \text{eq 1}$$

Where, ADI = average As intake ($\mu\text{g kg}^{-1}\text{-BW day}^{-1}$), C = As concentration in water, rice and other food ($\mu\text{g L}^{-1}$ for water, $\mu\text{g kg}^{-1}$ for food), IR = ingestion rate (L day^{-1} or g day^{-1}), EF = exposure frequency (days yr^{-1}), ED = exposure duration (years), BAV = bioavailability of As (unitless), BW = average body weight of the receptor (kg) and AT = averaging time of the exposure (days) [ED x 365, life expectancy x 365]. The total As ADI for each individual was equal to the sum of each individual ADI for each food and water component.

2.5 Estimation of non-carcinogenic and carcinogenic risk

Since analyzing individual level dietary intake from individual exposure pathways provides a good basis for investigating the influence of different parameters and exposure pathways on risk, the Hazard Quotient (HQ) and cancer risk were estimated for each study population separately using the individual level dietary intakes from water, rice, vegetables, protein and dal.

2.5.1 Chronic (non-cancer) risk

The extent of non-carcinogenic harm incurred due to exposure is referred to as non-cancer risk (Lee et al., 2005) and was estimated by calculating the Hazard Quotient (HQ) from exposure of As in water, rice and other foods (equation 2).

$$\text{HQ} = \frac{\text{ADI}}{\text{RfD}} \quad \text{eq 2}$$

Where ADI = average daily intake dose of a chemical ($\text{mg kg}^{-1} \text{day}^{-1}$) and RfD = reference dose ($\text{mg kg}^{-1} \text{day}^{-1}$). The reference dose is the daily chemical dose that results in no long-term harmful health effects from prolonged exposure (Lee et al., 2005) and was taken to be $3 \times 10^{-4} \text{ mg kg}^{-1} \text{day}^{-1}$ as derived from the US EPA IRIS (Integrated Risk Information System) toxicological database. HQ is thus a measure of relative toxicity, which compares actual exposure incurred from dietary intake of As (ADI) with the RfD for As. Individual HQ values for water and various foods when aggregated derive the hazard index (HI) which is a measure of overall chronic risk.

2.5.2 Carcinogenic risk

Carcinogenic risk (CR) from a given chemical, defined as the probability of developing cancer at a given lifetime exposure level (Lee et al., 2005), was calculated using equation 3 (Paustenbach, 2002).

$$\text{Cancer Risk} = \text{ADI}_{\text{life}} \times \text{SF} \quad \text{eq 3}$$

Where ADI_{life} = Life time exposure level and SF = Slope factor. The slope factor corresponds to the slope of the dose-response curve in the low dose region where the relationship between intake of chemical ($\text{mg kg}^{-1} \text{BW day}^{-1}$) and probability of developing cancer is assumed to be linear (Lee et al., 2005). The value adopted here was 1.5 as SF for As, as noted in the IRIS database (USEPA, 2003). Therefore, SF is the probability of developing cancer per unit of As exposure in $\text{mg kg}^{-1} \text{BW day}^{-1}$ (Lee et al., 2005). The lifetime exposure level (ADI_{life}) was derived by extrapolating the exposure burden over the life expectancy for the Bangladeshi male and female population.

2.6 Database management and mapping

Knowledge of the spatial variability of ADI, HQ and CR is important for the efficient management of contaminated landscapes. Complex database management (linking, querying) was performed to quantify the individual ADI, HQ and CR for the exposed population. The whole process of ADI, HQ and CR calculation (equation 1-3) was performed using Microsoft Access. Laboratory measured As concentrations

in water and food were structured in Microsoft Excel and subsequently, imported to the Access database. Arsenic exposure through water, rice, vegetables, dal and protein by adult males, adult females and children were calculated separately to identify the relative contribution of different food types to the total As intake as well as age and gender specific quantification of As exposure. The ADI, HQ and CR datasets were linked with the household feature class in ArcGIS to perform GIS analysis and mapping.

3 Results and Discussions

3.1 Gender and age specific daily intake of water and food

Differences in intake patterns were identified by dividing the population (n=345) into three categories based on age and gender; child (age ≤ 13 years), adult male (age > 13 years, gender = male) and adult female (age > 13 years, gender = female). Regardless of age or gender, water and rice dominated dietary composition (Figure 1). The median water intake was 2.5 L person⁻¹ day⁻¹ (range 0.2 – 7.0 L person⁻¹ day⁻¹; n=345) where the gender and age specific daily water intake are presented in Table 1. The median rice consumption was 1260 g person⁻¹ day⁻¹ (range 50 – 4133 g FW person⁻¹ day⁻¹; n=345) with the gender and age specific daily rice intake presented in Table 1. Total daily intake of vegetables irrespective of age and gender was 133 g FW person⁻¹ day⁻¹. The WHO recommends that a vegetable intake of 200 g FW vegetables person⁻¹ day⁻¹ is required to maintain good nutrition (Hassan et al., 2000). Comparison of the current study estimates with the WHO guideline suggests that the study population from all villages consumed significantly less vegetables than recommended. The average daily dry weight (DW) dal consumption was 171 g⁻¹ day⁻¹.

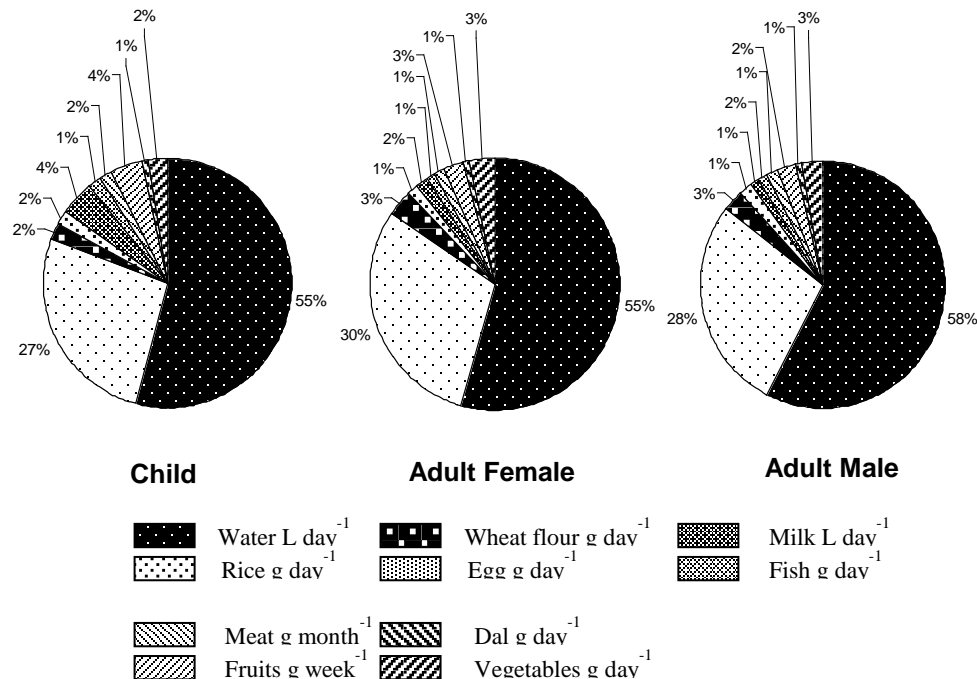


Figure 1 Age and gender specific relative contributions of water, rice and other foods to the average daily rural diet in the Sirajdikhan Upazila.

Table 1 Variation of average daily water intake ($L \text{ person}^{-1} \text{ day}^{-1}$) and daily cooked rice intake ($FW \text{ g person}^{-1} \text{ day}^{-1}$) by child, adult female and male with villages.

Village	Water Intake ($L \text{ person}^{-1} \text{ day}^{-1}$)			Rice Intake ($FW \text{ g person}^{-1} \text{ day}^{-1}$)		
	Child	Adult Female	Adult Male	Child	Adult Female	Adult Male
Daniapara	0.9 (n=11)	2.3 (n=24)	3.0 (n=20)	466 (n=11)	1371 (n=24)	1500 (n=20)
Abirpara	1.4 (n=18)	2.5 (n=30)	2.8 (n=22)	842 (n=18)	1647 (n=30)	1757 (n=22)
Dakatipara	1.9 (n=10)	2.9 (n=21)	3.3 (n=23)	647 (n=10)	1527 (n=21)	1798 (n=23)
UttarTajpur	1.9 (n=9)	2.6 (n=24)	3.2 (n=23)	822 (n=9)	1300 (n=24)	1525 (n=23)
Rashunia	1.4 (n=18)	2.5 (n=19)	3.5 (n=19)	751 (n=18)	1590 (n=19)	1787 (n=19)
Tenguripara	1.3 (n=15)	3.4 (n=21)	4.4 (n=18)	586 (n=15)	1271 (n=21)	1358 (n=18)
Overall average	1.4 (n=81)	2.7 (n=139)	3.3 (n=125)	697 (n=81)	1457 (n=139)	1628 (n=125)

3.2 Arsenic concentration in water, rice and other foods

The overall mean As concentration in tubewell (TW) water across all six villages was $48 \mu\text{g L}^{-1}$ and ranged between 0 to $808 \mu\text{g L}^{-1}$. Arsenic in TW water also varied significantly between households even when within close proximity ($<50 \text{ m}$). As concentrations in TW water exceeded the WHO guideline of $10 \mu\text{g L}^{-1}$ As and the Bangladesh guideline of $50 \mu\text{g L}^{-1}$ As, 73% and 10% of the time, respectively. The overall mean As concentration in rice grain was 0.151 (range 0.077 - 0.120 ; $n=60$) $\mu\text{g g}^{-1}$ DW. The mean As concentration in cooked rice was 0.034 (range 0.012 – 0.120 ; $n=60$) $\mu\text{g g}^{-1}$ FW. While some previous studies have reported elevated levels of As in cooked rice (Bae et al., 2002; Rahman et al., 2006; Ohno et al., 2007) in this study As concentrations in rice grain were much higher than in cooked rice. In many of the previous studies, rice was cooked with As contaminated water, whereas in this study cooking did not elevate As concentrations due to the use of As free pond water. The overall mean As concentrations in vegetables were 0.015 (range 0 - 0.136 ; $n=733$) $\mu\text{g g}^{-1}$ FW.

3.3 Gender and age specific average daily intake of As (ADI) from water, rice and other foods

While there was no variation in the frequency of consumption of either water or food with gender, there were significant differences in the amount of food and water consumed with gender. Adult males generally consumed relatively larger portions of food than adult females, and children consumed relatively smaller amounts than the adults. Determination of gender specific As daily intake was important because As had previously been shown to affect men and women differently (Heck, 2006). In the present study, although villagers had switched to pond water for cooking purposes, more than 73% of the surveyed households still obtained their drinking water from highly contaminated tubewells.

The overall average dietary As intake (ADI) via water was $2.10 \mu\text{g kg}^{-1} \text{ BW day}^{-1}$ ($n=345$), and 3.44 , 1.42 and 1.97 for the child, adult female and adult male populations respectively. The child population was highly exposed to As through drinking water due to the high As concentrations in water and their relatively lower body weight despite their lower daily water intake. Middle aged people (25-50 years) were also highly vulnerable to As exposure through water ingestion. There was a negative correlation between As intake through water and age group for both male ($r=-0.56$) and female ($r=-0.41$) populations. Of the total surveyed adult population, 34% exceeded the WHO's provisional maximum tolerable daily intake of As (MTDI) of $2.1 \mu\text{g kg}^{-1} \text{ BW day}^{-1}$ and among these, 21% were female and 13% male. A significant linear relationship was observed between ADI and the concentration of As in water ($r = 0.86$).

The overall average dietary intake of total As (TAs) through rice was $0.89 \mu\text{g kg}^{-1} \text{ BW day}^{-1}$ FW (range 0.07 to 5.38 ; $n=345$). Assuming an inorganic As content of 80% (Williams et al., 2006), the overall average dietary intake of inorganic As (IAs)

through rice was $0.71 \mu\text{g kg}^{-1} \text{ BW day}^{-1} \text{ FW}$ (range 0.06 to 4.31; $n=345$) and contributed 42% to the MTDI. The dietary intake of rice from total As (2.4 times lower) and inorganic As (2.95 times lower) are both lower than the WHO's MTDI of $2.1 \mu\text{g kg}^{-1} \text{ BW day}^{-1}$, which is also 2.4 times less than average dietary intake of As via water. Both household and village level dietary intake of As via rice indicated that there was a significant relationship between cooked rice As concentration ($R^2 = 0.74$ and $R^2 = 0.85$) and dietary intake of As, while there was no significant relationship ($R^2 = 0.15$ and $R^2 = 0.21$) between As dietary intake and the amount of rice consumed. It is evident from the relationship between As concentrations in cooked rice and mean household dietary intake of As that the study populations are not exceeding the MTDI As values by ingesting rice alone. There was a negative linear relationship between ADI of As from rice ingestion and body weight for both male ($r = -0.61$) and female ($r = -0.48$) populations indicating that people with a lower body weight are more vulnerable to As dietary exposure.

The relative contributions of water, rice and other foods to total dietary As intake were 26, 72 and 2%, respectively indicating dietary intake of As was dominated by water and rice. Across the study area As intake via water was related to the As concentration in the TW water and where As concentrations in TW water were low there was a consequential increase in the percentage As intake via rice. Therefore, in Sirajdikhan the relative contribution to As intake from rice was relatively high (72%) due to relatively low As concentration in TW water.

The dietary intake of As varied spatially across the villages, and in general corresponded to the variation in As concentrations in the household TW water and the regional As contamination level. However, significant spatial variability existed, even within short distances across a village. The regional variation of household level dietary intake of As from all sources ranged from 0.22 to $41.35 \mu\text{g kg}^{-1} \text{ BW day}^{-1}$. Highly exposed households were located in Abirpara and Tenguripara and dietary As exposure increased towards villages located in the south and south-east of Sirajdikhan (Figure 2)

3.4 Gender and age specific non-carcinogenic risk from water, rice and other food

The hazard quotient from water ingestion (HQ_W) was 7.23 ($n=345$) and contributed significantly (26%) to the Hazard Index (HI). Regression analysis indicated that when As concentrations in drinking waters were very high, the amount of water intake was not a significant contributor to risk ($R^2 = 0.006$, $p = 0.284$). Therefore, the non-carcinogenic risk from As exposure can be primarily minimized by introducing alternative water sources (or by removing As from the water) rather than by decreasing the amount of water ingested. The variation of HQ_W with age and gender, indicated that the risk level was higher for the child compared with the adult population. Risk gradually decreased with age due to body weight and relatively lower amounts of food intake compared with the younger adult population. The hazard quotient from rice ingestion (HQ_R) was 2.76 ($n=345$) and the percentage contribution of HQ_R to HI was 72% in agreement with the findings that "where As concentration in drinking water was lower the relative contribution of risk from rice ingestion was higher compared with water ingestion". To determine the statistical significance of the variation of HQ_R between villages ANOVA was performed and indicated that risk varied significantly between the villages ($R^2 = 0.80$, $p < 2.2e^{-16}$). Correlation coefficient tests also identified, that both As concentration in cooked rice and the amount of ingested cooked rice had a significant influence on HQ_R , but As concentrations in cooked rice ($r = 0.87$, $p < 2.2e^{-16}$) contributed more to HQ_R than the amount of ingested cooked rice ($r = 0.39$, $p = 8.498e^{-8}$). The HQ_R values from dietary intake of rice also varied significantly between households ($R^2 = 0.62$, $p < 2.2e^{-16}$) and with age and gender due to variations in body weight and the amount of cooked rice ingested.

3.5 Gender and age specific carcinogenic risk from water, rice and other food

The overall carcinogenic risk from water ingestion ($Risk_W$) was 2.22×10^{-3} , contributing 26% to the total risk. Cooked rice was identified as the second highest contributor towards risk of adverse health behind water with an overall $Risk_R$ of 7.59×10^{-4} ($n=345$). All villages showed significant differences in risk levels from ingestion of cooked rice ($R^2 = 0.58$, $p < 2.2e^{-16}$). Arsenic concentration ($R^2 = 0.63$, $p < 2e^{-16}$) in cooked rice had a higher contribution to risk compared to the amount cooked rice intake ($R^2 = 0.10$, $p = 4.7e^{-4}$). $Risk_R$ also varied significantly between different age groups ($r = -0.30$, $p < 2.2e^{-16}$) with a negative correlation that indicated $Risk_R$ decreased as age increased. None of the study population exceeded the acceptable level of risk through vegetable dietary exposure. The overall estimated cancer $Risk_V$ was 1.14×10^{-5} and $Risk_V$ only contributed 1.8% to the total risk.

Maps of lifetime risk due to dietary As intake showed that risk varied spatially between households within a village even within close proximity (< 100 m) and that elevated risk corresponded to higher groundwater As concentrations. The lifetime human health risk due to dietary exposure to As also varied significantly between villages located within the same landscape (Figure 2).

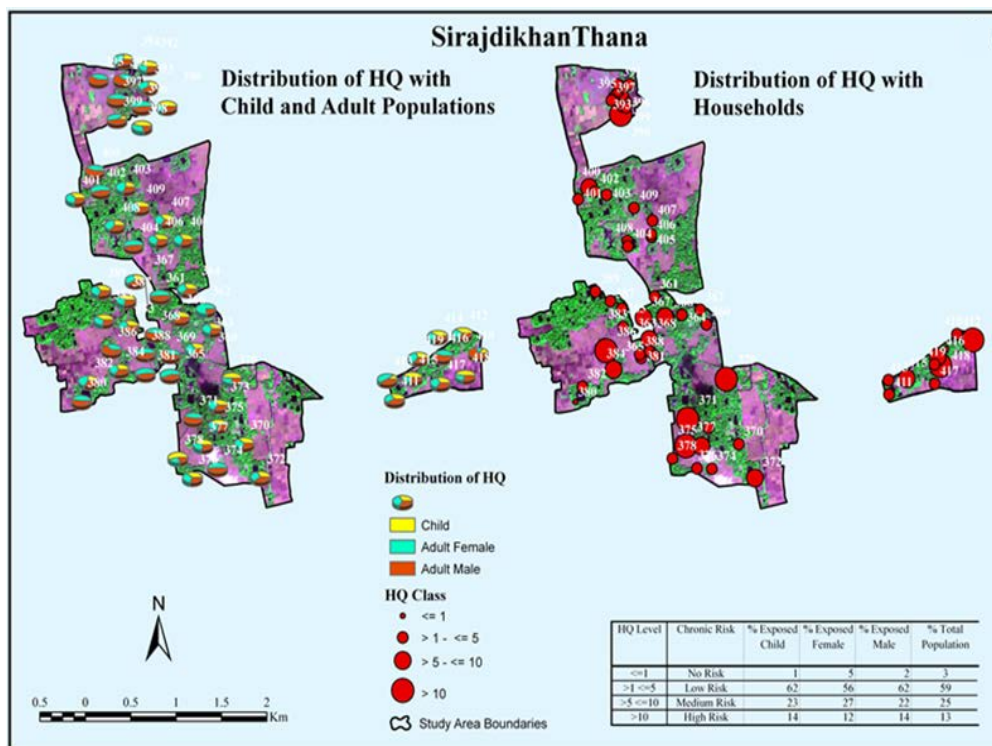


Figure 2 Spatial distribution of household chronic As risk through direct water and food ingestion pathways among child and adult populations in Sirajdikhan upazila overlaid on a Quickbird satellite image.

4 Conclusion

Dietary pattern and daily intake of food are vital inputs for accurate assessment of exposure, health risk and nutritional status. This study showed that the rural Bangladeshi diet was simple and homogeneous and typically consisted of a large portion of rice ($1340 \text{ g FW day}^{-1}$) with vegetables ($136 \text{ g FW day}^{-1}$) and dal (35 g FW day^{-1}), supplemented by the irregular consumption of fish or other meat protein. Beverages were not elaborate and routinely involved consumption of water alone (2.6 L day^{-1}). Therefore rice was the major source of protein. From the relative contribution of different foods to the daily diet this study concluded that water and rice were the major dietary exposure pathways for the rural population due primarily to their larger intake amounts and higher frequency of consumption irrespective of

As concentration. Dietary exposure through water ingestions was a more substantial contributor to the total daily As intake than any other food, especially at the higher levels of As concentration in tubewell water. Only when As concentrations in tubewell waters was low would rice become a substantial contributor to dietary As intake. Vegetables and dal contributed only 1% to the total dietary As intake. The lifetime risk (non-carcinogenic and carcinogenic) was mainly due to dietary intake of As through water and rice. The increased lifetime human health risk was closely associated with the As concentrations in drinking water and the contribution of As via dietary intake of vegetables, protein and dal was negligible, indicating that with the exception of rice, further extensive dietary surveys of other food items are not warranted. Human health risk can most efficiently be reduced by introducing alternative sources of As free drinking water.

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