

Health Risk Assessment for Bangladesh is due to Arsenic Exposure from Consumption of Vegetables Grown with Natural Arsenic Contaminated Groundwater

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Abstract

In the context of increasing uses of Arsenic contaminated groundwater for irrigation in Bangladesh and observed health related problems, we evaluated potential health risk for Bangladesh is due to As exposure from consumption of vegetables. Eight vegetable plants (Amaranth, Arum, Carrot, Eggplant, Indian Spinach, Okra, Potato and Radish) were grown in open field and irrigated with natural As contaminated water (0.005-0.903 mg L⁻¹) for 80-90 days. The average As concentrations in the vegetables, measured by GF-AAS, were 0.431, 0.719, 0.928, 1.574 and 2.287 mg kg⁻¹ (ww.) corresponding to irrigation water As concentrations of 0.005 (control), 0.044, 0.103, 0.507 and 0.903 mg L⁻¹ respectively. We established linear regression equation of fitted model for water-vegetable As concentration (n=54) relationship (r=0.767 and P<0.05). Statistical analyses with r value, P value of ANOVA table, Durbin-Watson Statistic and Lack-of-Fit test strongly validated the model. Merging British Geological Survey's (BGS) groundwater datasets (n=3534) for Bangladesh to the model led to estimation of As concentrations in vegetables district-wise. We followed USEPA Guidelines for Exposure Assessment for evaluation of human health risk. Risk, defined as 'Hazard Quotient' (HQ), was mapped for three vulnerable population sub-groups: Highly Exposed Child, Average Person and Senior. The results showed that all the children (0-6 years) were at health risk, whereas 98% of seniors and 76% of average persons (i.e., adults) were safe in consuming vegetables. The eight administrative division-wise HQ values for average persons were as follows: Rangpur (0.760)<Rajshahi (0.775)<Mymensingh (0.805)<Barisal (0.815)<Sylhet (0.820)<Khulna (0.990)<Dhaka (0.996)<Chittagong (1.147). The worst affected district was Chandpur. The findings explored the extent of As health risk for children (0-6 years), adults and seniors of Bangladesh precisely from vegetable consumption. This study highlights the importance of sight-specific risk assessment considering more pollutant parameters.

Keywords: Human health; Risk assessment; Exposure; Groundwater; Vegetables

Abbreviations:

As: Arsenic; HQ: Hazard Quotient; BGS: British Geological Survey; WHO: World Health Organization; BW: Body Weight; IR: Ingestion Rate; ADD: Average Daily Dose; DDW: Distilled De-Ionized Water; NOAEL: No Observed Adverse Effect Level; LOAEL: Low Observed Adverse Effect Level; RfD: Reference Dose; CRM: Certified Reference Materials.

Introduction

Inorganic Arsenic, As (III) and As (V), has been classified as a human carcinogen of Group 1 by International Agency for research on Cancer [1]. A study conducted by the BGS reported that the groundwater (n=3540) of shallow aquifer, used for both drinking and irrigation purposes, in 61 out of 64 districts of Bangladesh was contaminated with As [2]. In Chapai Nawabganj district of Bangladesh the highest concentration of As in tube-well water was found as 2,400 µg L⁻¹ [2], whereas WHO guideline value for drinking water was 50 µg L⁻¹ for Bangladesh and 10 µg L⁻¹ for many other countries [3]. An estimated 24 million people are directly exposed to this contamination and another 75 million are at risk [4,5]. Nearly 10,000 Arsenicosis

patients had been identified and a few deaths due to As related diseases had also been reported [4,6-8]. Evidently, Bangladesh is facing probably the largest mass poisoning in history.

The area under irrigation has been increased significantly over the last two decades in Bangladesh to raise food production mainly through installation of shallow tube-wells [9]. The As accumulated in soil due to irrigation is bio-transferred to vegetable and corn plants. It was reported [10,11] that Arum, Amaranth and Ipomoea were good As accumulators, with the levels of As in Arum and Potato up to 153 and 2.4 mg kg⁻¹ (dw.) respectively. They found As concentration in irrigation water in affected areas in the range 0.136-0.555 mg L⁻¹. Islam [12] found that As accumulated in vegetables exponentially due to successive harvesting. Obviously, increased use of As contaminated groundwater for irrigation purpose suggests that consumption of the irrigated crops and vegetables could be another major exposure route of As in humans in Bangladesh.

Irrespective of exposure pathways, As is a cumulative poison that does not metabolized in other intermediate compounds easily and does not easily break down in the environment [13]. As causes acute lethality to chronic effects including vascular diseases, hypertension, cancer, hyperpigmentation, genotoxicity, diabetes mellitus, repeated abortions, stillbirth, preeclampsia, etc. [5,14,15].

Since As problem in Bangladesh is of great concern at present, it is very important to understand the entrance of As into human food

chain from sources other than contaminated drinking water e.g., from consumption of vegetables. The health threat might be increased to many-folds due to high abundance of As in the earth's crust of Bangladesh [2] as well as use of Arsenical pesticides and sludge, and repeated irrigation with contaminated water. It is predicted that in Bangladesh 200,000-270,000 people will die of cancer by drinking As contaminated water alone [16]. But in Bangladesh limited studies were performed for integrated human health risk assessment. For example, Islam [17] from market basket survey from Bogra region of Bangladesh for As evaluated Target Hazard Quotient (THQ) and found increased risk of cancer for the concerned people.

Our investigation aimed to evaluate human health risk for different vulnerable population sub-groups of society (Highly Exposed Child, Average Person and Senior) of Bangladesh from As exposures due to consumption of different types of vegetables. For this, we estimated total As content in vegetables (by GF-AAS) cultivated with natural As contaminated groundwater, developed statistically verified water-vegetable As concentration relationship and merged the data of BGS (n=3540) to get predicted As concentrations of vegetables district-wise. Following USEPA Guidelines for Exposure Assessment [18,19] we estimated ADD and HQ, and generated three HQ maps. This study will help to assess the non-carcinogenic health risk for Bangladesh is with regard to consumption of both aboveground and belowground vegetables for young children (0-6 years), adults and seniors.

Materials and Methods

Soil preparation, seeding and irrigation

The studied eight vegetables (Amaranth, Arum leaf, Carrot, Eggplant, Indian Spinach, Okra, Potato and Radish), both aboveground and belowground, were cultivated in an open field that was under an agro-ecological zone of the Ganges Fluvial Floodplain. The location of the field was 24°22'25.14" N and 88°37'41.98" E. The land was divided into 5 × 8 beds, each of area 1 ft × 1 ft. Each bed was separated from another by 1½ ft embankment having height of 1 ft to prevent mixing of irrigation water and seepage. With fresh soil commercially available urea, TSP and potash were mixed well in the N-P-K ratio of 2:1:1 along with cattle manure and insecticide carbofuran [20]. The soil was allowed to stand for at least 120 h to stabilize the mix.

After seeding, the beds were watered with natural groundwater having As concentrations of 0.005, 0.044, 0.103, 0.507 and 0.903 mg L⁻¹. The groundwater were collected from shallow aquifer of Rajshahi, Bangladesh through tube-wells (location and characteristics are mentioned in SI). Groundwater having Ar concentration of 0.005 mg L⁻¹ was applied as control run. The plants of interest were watered 10 times over a period of 80-90 days with a usage of 3.0 L/bed/irrigation. The average interval of irrigation was 7 days that ensured field level moisture content in soil.

Sampling

Prior to water sample analysis, the tube-wells were flushed with 2-3 tube-well volumes of groundwater (e.g., 70 L for tube-well having depth of 20 m and internal diameter of 4 cm) [21,22]. The clear water samples were collected in 500 ml acid-washed polyethylene bottles, acidified with 69% HNO₃ to keep pH below 2 and stored in refrigerator at 4°C [23]. For edible plant tissue sampling, the vegetables were rinsed with DDW three times, cut into pieces, dried in an oven at

70°C for 48 h and ground in a ceramic mortar followed by sieving with 0.5 mm screen. The soils were collected from 0-6 cm beneath the surface layer and a distance from the root in polyethylene bags. The soils were then dried in an oven at 105°C for 24 h, ground and sieved with 0.5 mm screen.

Digestion and analysis

For extraction of As, the water samples were subjected to mild digestion with HNO₃ [23], soil samples were digested by modified method of Small and McCants with H₂SO₄-HClO₄ (volume ratio of 2:3) [24-26] and vegetable samples were digested through Wet Oxidation by means of ternary acid mixture HNO₃-H₂SO₄-HClO₄ (volume ratio of 10:1:4) [25-27]. Prior to wet oxidation the samples were pre-digested with 69% (v/v) HNO₃. The digestates, after dilution with 6 M HCl and DDW, were analyzed for total concentrations of As by GF-AAS using a Shimadzu AA-6800 (Shimadzu Corporation, Kyoto, Japan) atomic absorption spectrophotometer. It was equipped with an auto-sampler (ASC-6100, Shimadzu) and a graphite furnace (GFA-EX7, Shimadzu), and operated through 'WizAArd' software. The minimum detection limit of As was 0.3 µg L⁻¹. Other parameters were also measured for cultivated soil and groundwater according to standard procedures.

Model development

$$As_{ADD} = \frac{As_{veg} \cdot IR \cdot ED}{BW \cdot AT} \quad (1)$$

For estimating total exposure of As for the consumers of vegetables, we first estimated ADD (mg kg⁻¹ day⁻¹) using equation (1) as suggested by USEPA Guidelines for Human Exposure Assessment [18,19].

Where As_{veg} = average concentration of As in the vegetables (mg kg⁻¹, dw.), IR=Ingestion Rate for vegetables (kg day⁻¹, ww.), ED=Exposure Duration (yr), BW=Body Weight (kg) and AT=Averaging Time (yr). We considered bio-accumulation by multiplying equation (1) by Relative Bio-availability Factor, RBAF. This along with vegetable Conversion Factor, CF_{veg} (dry weight to fresh weight) leads to,

$$As_{ADD} = \frac{As_{veg} \cdot CF_{veg} \cdot IR \cdot ED}{BW \cdot AT} \times (RBAF) \quad (2)$$

In Bangladesh, there is no comprehensive datasets for As concentrations in vegetables that consumed countrywide; only some discrete surveys were made. BGS of UK and Department of Public Health Engineering (DPHE) of Bangladesh [24] made an extensive national hydro-chemical survey across Bangladesh. We utilized their groundwater As datasets (n=3534) to obtain As concentrations in vegetables countrywide. Our experimentally established linear regression equation of the fitted model for water-vegetables As content (n=54) (Figure 1), verified well statistically, was utilized for the purpose.

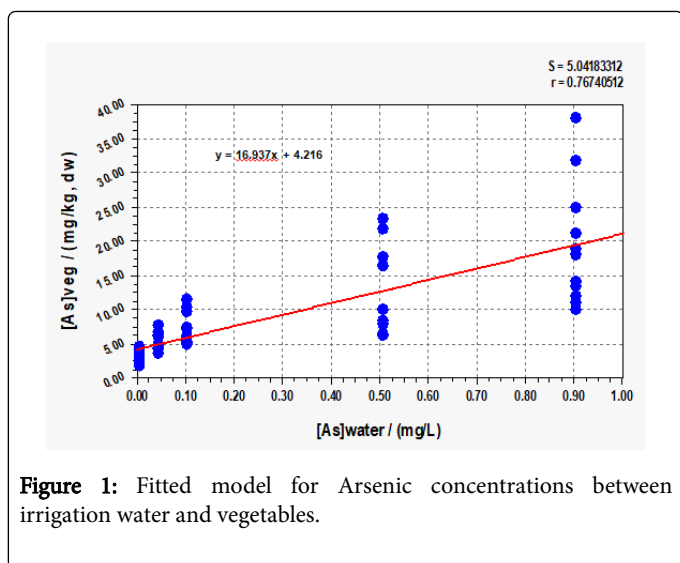


Figure 1: Fitted model for Arsenic concentrations between irrigation water and vegetables.

The National Nutrition Survey (NNS) [28], Household Expenditure Survey (HES) [29] and other sources [30] showed that cereals represented the largest amount of food consumed (436 g/caput/day) followed by fruits and vegetables (126 g/caput/day) and, roots and tubers (72 g/caput/day). Fruit consumption being very small, we considered 200 g/person/day as IR of vegetables for Average Person group. We assumed the IR values for child of age 6 years and senior as 65 and 100 g/person/day respectively. The consumption level is fairly low with respect to WHO recommended value of vegetables and fruit intake, which is 400 g/person/day [31]. The other parameters involved in ADD estimation are summarized in Table 1. These were adapted from the data of Exposure Factor Handbook [32], USEPA standard values [33] and Bangladesh Surveys and Standards [29]. ADD were estimated separately for three vulnerable population sub-groups: 'Highly Exposed Child', 'Average Person' and 'Senior' (Table 1).

| Factors | Unit | Population sub-group | | |
|---|--------------------------|----------------------------------|----------------|--------|
| | | Highly Exposed Child (0-6 years) | Average person | Senior |
| Age | yr | 6 | 39.5 | 60.3 |
| Body Weight (BW) | kg | 18.6 | 80.8 | 82.6 |
| Vegetable ingestion rate | g (ww) day ⁻¹ | 65 | 130 | 100 |
| Exposure Frequency (EF) | day yr ⁻¹ | 350 | 350 | 350 |
| Year | day | 365.25 | 365.25 | 365.25 |
| Averaging Time (AT) | yr | 6 | 30 | 50 |
| Exposure Duration (ED) | yr | 6 | 30 | 50 |
| Relative Bio-availability Factor (RBAF) for Arsenic | Unit less | 0.4 | 0.4 | 0.4 |
| Conversion Factor (Fresh wt. to Dry wt.) (CF) | Unit less | 0.085 | 0.085 | 0.085 |

Table 1: Parameters involved in the risk assessment model.

Risk characterization

Risk may be characterized using a HQ. This is the ratio of ADD of a chemical to a RfD defined as the maximum tolerable daily intake of a specific metal that does not result in any deleterious health effects. The RfDs may be derived from a NOAEL or a LOAEL were adapted from USEPA's Health Effects Notebook and Exposure Factors Handbook [32] and California EPA's RfD values [34]. For As,

$$HQ = \frac{As_A DD}{As_R fD} \quad (3)$$

Integrated Risk Information System (IRIS) of USEPA [35] has set the RfD for inorganic As as $3.0 \times 10^{-4} \text{ mg kg}^{-1} \text{ day}^{-1}$ based on hyperpigmentation, keratosis and possible vascular complications in humans (from NOAEL and LOAEL values of 8.0×10^{-4} and $0.014 \text{ mg kg}^{-1} \text{ day}^{-1}$ respectively). If $HQ > 1.000$, then the ADD of a particular metal exceeds the RfD indicating that there is a potential risk associated with that metal.

Quality control

All glassware were treated with 10% (v/v) HNO₃ for 24 h and then rinsed three times with DDW followed by drying in an oven. CRM of As standard solutions for AAS (TraceCERT[®]) (Fluka, Switzerland) were used for calibration purposes. An analysis was carried out following USEPA approved Quality Assurance/Quality Control (QA/QC) plan with a reagent blank, a duplicate and a spike for every 20 samples. After analyzing every 10 samples, readings of standard solutions were recorded to check the instrument. Two CRM (Tomato Leaves (SRM 1573a) and Lake Sediment (NIES 31)) and one internal reference material (prepared very cautiously by control water with CRM standard solution for AAS) were digested and analyzed in five replicates for total As concentration under the identical experimental conditions to check accuracy of the method. The overall agreement between the certified and measured values was 97-104% (Table 2 (Supplementary Information)).

| Sample | Certified values (mg kg ⁻¹) | Measured values ^a (mg kg ⁻¹) | % of recovery | n ^b |
|-----------------------------------|---|---|---------------|----------------|
| Tomato Leaves (SRM 1573a) | 0.112 ± 0.004 | 0.117 ± 0.009 | 104 | 5 |
| Lake Sediment (NIES CRM No. 31) | 13.9 ± 1.5 | 13.5 ± 1.3 | 97 | 5 |
| Groundwater (Prepared internally) | 0.100 ± 0.11 ^c | 0.103 ± 0.08 ^c | 103 | 4 |

Table 2: Analysis of certified reference materials for total Arsenic concentration. Where, ^a=the samples were analyzed by GF-AAS, ^b=No. of measurements, ^c=unit was mg L⁻¹.

Statistical analyses

In our investigation each concentration value corresponds to an average of triplicate measurements. The datasets obtained were treated separately for analyzing basic statistical parameters and for making cross-tabulations and cross-plots. The SPSS (release 20.0), STATGRAPHICS Centurion (release 18.1.01) and Microsoft Excel (release 12.0) were employed for the purpose. Mathematical models were established based on simple and multiple regression analyses. The models were cross-checked by analyzing ANOVA, P value, r value (Pearson correlation coefficient), Durbin-Watson statistics and 'Lack-of-Fit' test. For these, Curve Expert (release 1.40) and STATGRAPHICS Centurion software were employed. The Box-Whisker plot was constructed using SPSS.

Results and Discussion

Distribution of Arsenic in vegetables

The statistical analyses of the observed concentrations of As (n=57) in the studied eight vegetables are presented in Table 3. Among the vegetables Amaranth (*Amaranthus* sp.), Arum (*Colocasia esculenta*) leaf, Eggplant (*Solanum melongena*), Indian Spinach (*Basella alba*) and Okra (*Ablemoschus esculentus*) are aboveground type and Carrot (*Daucus carota*), Potato (*Solanum tuberosum*) and Radish (*Raphanus sativus*) are belowground type.

| Vegetables | n | Concentration of Arsenic in vegetables ^a (mg kg ⁻¹) | | | | | | | | Dry wt.:Fresh wt. |
|----------------|----|--|-------------|--------|---------------|--------------------------------|--------------------|----------|------------------|-------------------|
| | | Control | Range | Median | Mean ± SE | 95% C.I. ^b for mean | Standard deviation | Variance | Skewness | |
| Amaranth | 11 | 0.399 | 0.273-5.431 | 0.912 | 1.383 ± 0.444 | 0.393-2.373 | 1.474 | 2.172 | 3.002 | 0.096 |
| Arum (Leaf) | 10 | 0.425 | 0.420-3.330 | 1.124 | 1.444 ± 0.305 | 0.754-2.133 | 0.964 | 0.930 | 1.145 | 0.067 |
| Carrot | 5 | 0.213 | 0.213-1.440 | 0.641 | 0.734 ± 0.203 | 0.171-1.296 | 0.453 | 0.205 | 0.322 | 0.104 |
| Eggplant | 5 | 0.384 | 0.384-4.046 | 1.060 | 1.970 ± 0.761 | -0.143-4.082 | 1.702 | 2.896 | 0.513 | 0.063 |
| Indian Spinach | 6 | 0.173 | 0.141-1.017 | 0.369 | 0.465 ± 0.135 | 0.118-0.812 | 0.331 | 0.109 | 0.693 | 0.070 |
| Okra | 10 | 0.345 | 0.253-1.539 | 0.789 | 0.830 ± 0.127 | 0.543-1.117 | 0.401 | 0.161 | -0.496 | 0.072 |
| Potato | 2 | 0.395 | 0.395-2.183 | 1.289 | 1.289 ± 0.894 | -10.070-12.648 | 1.264 | 1.598 | n/a ^c | 0.135 |
| Radish | 5 | 0.362 | 0.362-2.031 | 0.760 | 0.974 ± 0.290 | 0.168-1.779 | 0.649 | 0.421 | 1.255 | 0.122 |
| All | 54 | 0.341 | 0.141-5.431 | 0.812 | 1.143 ± 0.142 | 0.858-1.427 | 1.041 | 1.085 | 6.604 | 0.085 |

Table 3: Analysis of concentration of Arsenic in vegetables. Where, n=no. of observations; SE=Standard Error of mean; ^avalues in parenthesis=dry weight; C.I.^b=Confidence Interval; n/a^c=not applicable.

The observed concentrations of As in the edible plant tissues of vegetables, when irrigated with water of As lower concentrations, are in accordance with many authors [36,37]. The range of As concentrations in the vegetables were 0.141-0.436 (average 0.431), 0.346-1.168 (0.719), 0.392-1.446 (0.928), 0.689-3.565 (1.574) and 1.017-5.431 (2.287) mg kg⁻¹ (ww.) corresponding to irrigation water As concentrations of 0.005 (control), 0.044, 0.103, 0.507 and 0.903 mg L⁻¹ respectively. The As concentrations in the control vegetables lied within the Maximum Permissible Limits (MPL) of As in vegetables that are 0.5, 1.0, 1.4 mg kg⁻¹ (ww.) in China [38], Malaysia [39] and Hong Kong [40] respectively. It is to be noted that natural groundwater As concentration range of <1.000 mg L⁻¹ represented 99.94% of

groundwater of Bangladesh [2]. The distribution of As in edible plant tissues of the vegetables with respect to groundwater As concentrations is represented in the

Water-vegetable Arsenic concentration relationship

The linear regression equations of the fitted model for correlations of vegetables As concentration, [As]_{veg}, with irrigation water As concentrations was y=16.9374x+4.2166, where y=[As]_{veg} (mg kg⁻¹, dw.) and x=irrigation water As concentration (mg L⁻¹), against background soil As concentration of 9.935 mg kg⁻¹ (ww.). It is to be noted that the average As concentration in soil of Bangladesh was less

than 10 mg kg⁻¹ (ww.) [10]. The observed water-vegetables linear relationship (Figure 2) is important to get As concentrations in vegetables from applied irrigation water. Merging the groundwater As data (n=3534) of BGS/DPHE [2] to the regression equation led to of equation (2).

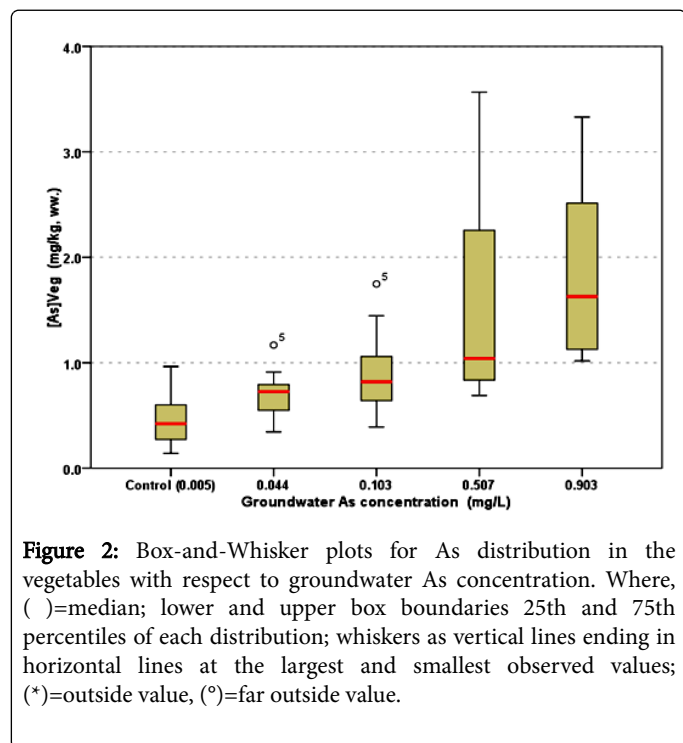


Figure 2: Box-and-Whisker plots for As distribution in the vegetables with respect to groundwater As concentration. Where, ()=median; lower and upper box boundaries 25th and 75th percentiles of each distribution; whiskers as vertical lines ending in horizontal lines at the largest and smallest observed values; (*)=outside value, (°)=far outside value.

In the model, r value of 0.767 indicated a moderately strong relationship between the variables. Since the P value of the ANOVA table was less than 0.05, there was a statistically significant relationship between the variables at the 95% confidence level. Since the Durbin-Watson value (1.53144) was greater than 1.4, there were no serious autocorrelation in the residuals. Moreover, analysis of variance with 'Lack-of-Fit' test revealed that the model was adequate for the observed data at the 95% confidence level. All the statistical analyses strongly validated our model.

Risk characterization

Risk assessment strategies are often aimed at population subgroups. It is common practice to identify vulnerable people in society, such young children or elderly, and assess potential risk to the health of these population subgroups [41]. Hough [42] considered young children to be Highly Exposed Individuals (HEIs). Thus risk assessment can usually focus on highly exposed sub-populations on the basis that if the risk to the HEI is acceptable then most of the population is protected.

The HQ is a screening risk assessment technique commonly used to judge whether there is concern for additive effects between chemicals. If the HQ is less than unity (1.000), it indicates that even if all the metals acted on the same organ or interacted in some other way to cause health effects, the risk of these effects would be low. The obtained ^{As}ADD values (mg kg⁻¹ day⁻¹) when divided by the ^{As}RfD, mg kg⁻¹ day⁻¹ (equation 3) yield the HQ values (Table 4). The estimated HQ values were utilized to generate HQ indexed maps for the three population sub-groups (Figure 3) showing clearly where the greatest potential risks were located.

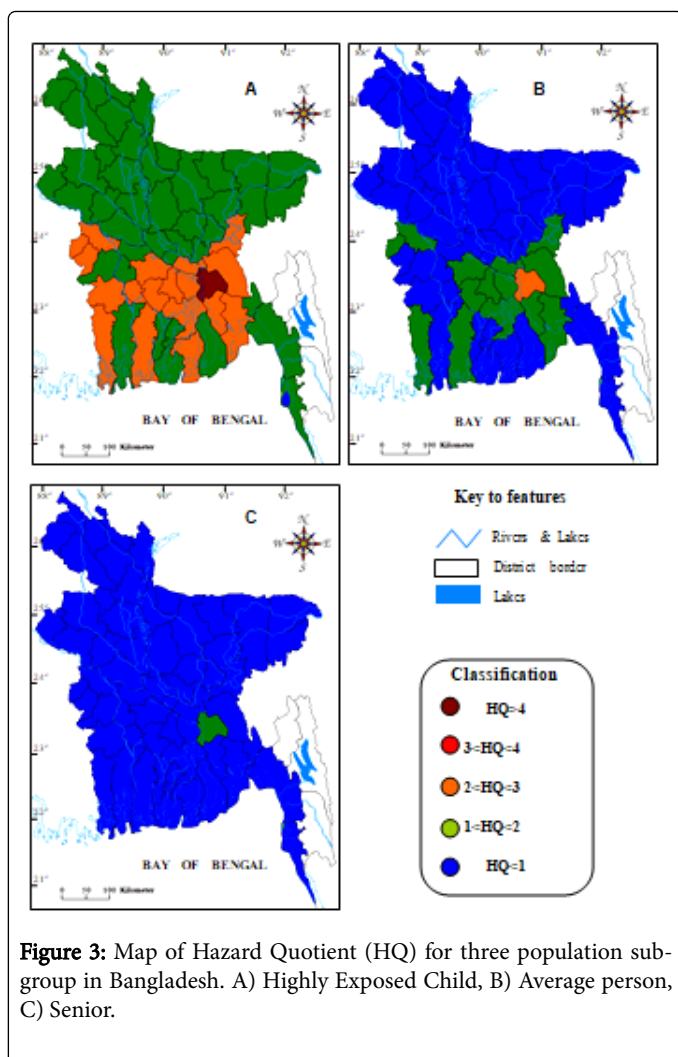
| District | Division | Child (0-6 yr) | | Average Person | | Senior | |
|------------------|----------|----------------|-------|----------------|-------|----------|-------|
| | | ADD | HQ | ADD | HQ | ADD | HQ |
| Bogra | Rajshahi | 5.15E-04 | 1.716 | 2.37E-04 | 0.79 | 1.78E-04 | 0.594 |
| Jaipurhat | Rajshahi | 4.82E-04 | 1.607 | 2.22E-04 | 0.74 | 1.67E-04 | 0.557 |
| Naogaon | Rajshahi | 4.92E-04 | 1.639 | 2.26E-04 | 0.754 | 1.70E-04 | 0.568 |
| Natore | Rajshahi | 4.82E-04 | 1.607 | 2.22E-04 | 0.74 | 1.67E-04 | 0.557 |
| Chapai Nawabganj | Rajshahi | 4.92E-04 | 1.639 | 2.26E-04 | 0.754 | 1.70E-04 | 0.568 |
| Pabna | Rajshahi | 5.42E-04 | 1.806 | 2.50E-04 | 0.832 | 1.88E-04 | 0.626 |
| Rajshahi | Rajshahi | 4.94E-04 | 1.645 | 2.27E-04 | 0.757 | 1.71E-04 | 0.57 |
| Sirajganj | Rajshahi | 5.40E-04 | 1.8 | 2.49E-04 | 0.829 | 1.87E-04 | 0.623 |
| Dinajpur | Rangpur | 4.86E-04 | 1.619 | 2.24E-04 | 0.746 | 1.68E-04 | 0.561 |
| Gaibandha | Rangpur | 5.23E-04 | 1.742 | 2.41E-04 | 0.802 | 1.81E-04 | 0.603 |
| Kurigram | Rangpur | 5.23E-04 | 1.742 | 2.41E-04 | 0.802 | 1.81E-04 | 0.603 |
| Lalmonirhat | Rangpur | 4.82E-04 | 1.607 | 2.22E-04 | 0.74 | 1.67E-04 | 0.557 |
| Nilphamari | Rangpur | 4.84E-04 | 1.613 | 2.23E-04 | 0.743 | 1.68E-04 | 0.559 |
| Panchagarh | Rangpur | 4.86E-04 | 1.619 | 2.24E-04 | 0.746 | 1.68E-04 | 0.561 |
| Rangpur | Rangpur | 4.96E-04 | 1.652 | 2.28E-04 | 0.76 | 1.72E-04 | 0.572 |

| | | | | | | | |
|---------------|------------|----------|-------|----------|-------|----------|-------|
| Thakurgaon | Rangpur | 4.82E-04 | 1.607 | 2.22E-04 | 0.74 | 1.67E-04 | 0.557 |
| Bagerhat | Khulna | 7.81E-04 | 2.603 | 3.60E-04 | 1.198 | 2.71E-04 | 0.902 |
| Chuadanga | Khulna | 6.33E-04 | 2.108 | 2.91E-04 | 0.971 | 2.19E-04 | 0.73 |
| Jessore | Khulna | 6.15E-04 | 2.05 | 2.83E-04 | 0.944 | 2.13E-04 | 0.71 |
| Jhenaidah | Khulna | 5.69E-04 | 1.896 | 2.62E-04 | 0.873 | 1.97E-04 | 0.657 |
| Khulna | Khulna | 5.48E-04 | 1.825 | 2.52E-04 | 0.84 | 1.90E-04 | 0.632 |
| Kushtia | Khulna | 6.81E-04 | 2.269 | 3.13E-04 | 1.045 | 2.36E-04 | 0.786 |
| Magura | Khulna | 5.34E-04 | 1.78 | 2.46E-04 | 0.82 | 1.85E-04 | 0.617 |
| Meherpur | Khulna | 7.04E-04 | 2.346 | 3.24E-04 | 1.08 | 2.44E-04 | 0.813 |
| Narail | Khulna | 6.50E-04 | 2.166 | 2.99E-04 | 0.997 | 2.25E-04 | 0.75 |
| Satkhira | Khulna | 7.37E-04 | 2.455 | 3.39E-04 | 1.13 | 2.55E-04 | 0.85 |
| Barguna | Barisal | 4.82E-04 | 1.607 | 2.22E-04 | 0.74 | 1.67E-04 | 0.557 |
| Barisal | Barisal | 6.58E-04 | 2.192 | 3.03E-04 | 1.009 | 2.28E-04 | 0.759 |
| Bhola | Barisal | 4.99E-04 | 1.665 | 2.30E-04 | 0.766 | 1.73E-04 | 0.577 |
| Jhalakati | Barisal | 5.24E-04 | 1.748 | 2.41E-04 | 0.805 | 1.82E-04 | 0.606 |
| Patuakhali | Barisal | 4.86E-04 | 1.619 | 2.24E-04 | 0.746 | 1.68E-04 | 0.561 |
| Pirojpur | Barisal | 5.38E-04 | 1.793 | 2.48E-04 | 0.826 | 1.86E-04 | 0.621 |
| Dhaka | Dhaka | 5.59E-04 | 1.864 | 2.57E-04 | 0.858 | 1.94E-04 | 0.646 |
| Faridpur | Dhaka | 7.50E-04 | 2.5 | 3.45E-04 | 1.151 | 2.60E-04 | 0.866 |
| Gazipur | Dhaka | 4.88E-04 | 1.626 | 2.25E-04 | 0.749 | 1.69E-04 | 0.563 |
| Gopalganj | Dhaka | 8.41E-04 | 2.802 | 3.87E-04 | 1.29 | 2.91E-04 | 0.971 |
| Kishorganj | Dhaka | 5.80E-04 | 1.934 | 2.67E-04 | 0.891 | 2.01E-04 | 0.67 |
| Madaripur | Dhaka | 8.49E-04 | 2.828 | 3.91E-04 | 1.302 | 2.94E-04 | 0.98 |
| Manikganj | Dhaka | 5.26E-04 | 1.755 | 2.42E-04 | 0.808 | 1.82E-04 | 0.608 |
| Munshiganj | Dhaka | 8.45E-04 | 2.815 | 3.89E-04 | 1.296 | 2.93E-04 | 0.975 |
| Narayanganj | Dhaka | 5.73E-04 | 1.909 | 2.64E-04 | 0.879 | 1.98E-04 | 0.661 |
| Narsingdi | Dhaka | 5.59E-04 | 1.864 | 2.57E-04 | 0.858 | 1.94E-04 | 0.646 |
| Rajbari | Dhaka | 5.75E-04 | 1.916 | 2.65E-04 | 0.882 | 1.99E-04 | 0.664 |
| Shariatpur | Dhaka | 7.71E-04 | 2.571 | 3.55E-04 | 1.184 | 2.67E-04 | 0.891 |
| Tangail | Dhaka | 5.19E-04 | 1.729 | 2.39E-04 | 0.796 | 1.80E-04 | 0.599 |
| Jamalpur | Mymensingh | 5.07E-04 | 1.69 | 2.34E-04 | 0.778 | 1.76E-04 | 0.586 |
| Mymensingh | Mymensingh | 5.11E-04 | 1.703 | 2.35E-04 | 0.784 | 1.77E-04 | 0.59 |
| Netrokona | Mymensingh | 5.57E-04 | 1.857 | 2.57E-04 | 0.855 | 1.93E-04 | 0.643 |
| Sherpur | Mymensingh | 5.23E-04 | 1.742 | 2.41E-04 | 0.802 | 1.81E-04 | 0.603 |
| Habiganj | Sylhet | 5.23E-04 | 1.742 | 2.41E-04 | 0.802 | 1.81E-04 | 0.603 |
| Moulavi Bazar | Sylhet | 5.19E-04 | 1.729 | 2.39E-04 | 0.796 | 1.80E-04 | 0.599 |

| | | | | | | | |
|---------------|------------|----------|-------|----------|-------|----------|-------|
| Sunamganj | Sylhet | 5.73E-04 | 1.909 | 2.64E-04 | 0.879 | 1.98E-04 | 0.661 |
| Sylhet | Sylhet | 5.23E-04 | 1.742 | 2.41E-04 | 0.802 | 1.81E-04 | 0.603 |
| Bandarban* | Chittagong | - | - | - | - | - | - |
| Brahmanbaria | Chittagong | 6.75E-04 | 2.249 | 3.11E-04 | 1.036 | 2.34E-04 | 0.779 |
| Chandpur | Chittagong | 1.32E-03 | 4.403 | 6.08E-04 | 2.027 | 4.58E-04 | 1.525 |
| Chittagong | Chittagong | 5.42E-04 | 1.806 | 2.50E-04 | 0.832 | 1.88E-04 | 0.626 |
| Comilla | Chittagong | 7.54E-04 | 2.513 | 3.47E-04 | 1.157 | 2.61E-04 | 0.871 |
| Cox's Bazar | Chittagong | 4.86E-04 | 1.619 | 2.24E-04 | 0.746 | 1.68E-04 | 0.561 |
| Feni | Chittagong | 5.84E-04 | 1.947 | 2.69E-04 | 0.897 | 2.02E-04 | 0.675 |
| Khagrachhari* | Chittagong | - | - | - | - | - | - |
| Lakshmipur | Chittagong | 8.25E-04 | 2.751 | 3.80E-04 | 1.266 | 2.86E-04 | 0.953 |
| Noakhali | Chittagong | 7.93E-04 | 2.642 | 3.65E-04 | 1.216 | 2.75E-04 | 0.915 |
| Rangamati* | Chittagong | - | - | - | - | - | - |

*Data was unavailable for the three districts of Chittagong Hill Tract.

Table 4: Average Daily Dose (ADD) and Hazard Quotient (HQ) for 60 districts of Bangladesh.



The HQ map (Figure 3C) reveals that 98% of the seniors were safe from consumption of vegetables, as $HQ < 1.000$. The average HQ for the entire senior population sub-group was 0.687. This suggests that almost all seniors would not experience any form of deleterious health effects due to living within and consuming vegetables in Bangladesh. It is evident from Figure 3B that the Average Person (i.e., Adult) population sub-group of 15 districts out of the studied 61 districts was at health risk for vegetables consumption. Amongst the 8 administrative divisions, Rangpur was least affected and Chittagong was worst affected for Average Person. For this population sub-group, the eight administrative division-wise HQ values were as follows: Rangpur (0.760) < Rajshahi (0.775) < Mymensingh (0.805) < Barisal (0.815) < Sylhet (0.820) < Khulna (0.990) < Dhaka (0.996) < Chittagong (1.147). The average value of HQ was 0.913. Inspection of district areas revealed that the Average Persons of 76% of the surveyed area, representing 69% of total area of Bangladesh, were safe in consuming vegetables.

Figure 3A reflects that all the Highly Exposed Children (age 0-6 years) were at health risk due to consumption of vegetables, since all exceeding HQ values of 1.000. This is attributed to relatively more consumption of vegetables with respect to their BW. The majority (70%) of the children had HQ values between 1.000 and 2.000. The average HQ for the Highly Exposed Child population sub-group was

1.983. Overall the human health safety order was as follows: Senior > Average Person > Highly Exposed Child. It is to be noted that for all population sub-groups, the worst affected district was Chandpur, where many rivers such as the Padma and the Meghna intersected, that experienced extensive river erosion. Mineralogical studies of the sediments showed the presence of As bearing minerals Iron arsenate ($Fe_2As(AsO_4)_3$) and As selenide telluride ($AsSe_{0.5}Te_2$) at significant levels in several depths [43]. Generally, the extent of such health risk is less in northern Bangladesh and more in southern Bangladesh.

In UK, similar safety order was observed by Hough [42] for consuming vegetables alone. BGS [2] reported that the groundwater of Bangladesh was also contaminated with Co, Mn, Mo, Rb, Sr, U, etc. Moreover, there are some 'Arsenic hot-spots' (a few kilometer across) that move under hydraulic gradient ($5-20 \text{ mm day}^{-1}$). Thus this study highlights the importance of sight-specific human health risk assessment considering more pollutant parameters.

Conclusion

In Bangladesh, the uses of As contaminated groundwater for irrigation are increasing day by day to raise food production and thereby creating wider As related health problems. We evaluated potential health risk for Bangladesh is due to As exposure from consumption of vegetables in this study. Eight vegetable plants (Amaranth, Arum, Carrot, Eggplant, Indian Spinach, Okra, Potato and Radish) were grown in open field and irrigated with natural As contaminated water ($0.005-0.903 \text{ mg L}^{-1}$) for 80-90 days. The average As concentrations in the vegetables were 0.431, 0.719, 0.928, 1.574 and 2.287 mg kg^{-1} (ww.) corresponding to irrigation water As concentrations of 0.005 (control), 0.044, 0.103, 0.507 and 0.903 mg L^{-1} respectively. We established linear regression equation of fitted model for water-vegetable As concentration ($n=54$) relationship ($r=0.767$ and $P < 0.05$). Statistical analyses with r value, P value of ANOVA table, Durbin-Watson Statistic and Lack-of-Fit test strongly validated the model. Merging BGS's groundwater datasets ($n=3534$) for Bangladesh to the model led to estimation of As concentrations in vegetables district-wise. We followed USEPA Guidelines for Exposure Assessment for evaluation of human health risk. Risk, defined as HQ, was mapped for three vulnerable population sub-groups: Highly Exposed Child, Average Person and Senior. The results showed that all the children (0-6 years) were at health risk, whereas 98% of seniors and 76% of average persons (i.e., adults) were safe in consuming vegetables. The HQ values for average persons of eight administrative divisions in Bangladesh were as follows: Rangpur (0.760) < Rajshahi (0.775) < Mymensingh (0.805) < Barisal (0.815) < Sylhet (0.820) < Khulna (0.990) < Dhaka (0.996) < Chittagong (1.147). The worst affected district was Chandpur. Generally the extent of such health risk is less in northern Bangladesh and more in southern Bangladesh. Thus the findings explored the extent of As health risk for children (0-6 years), adults and seniors of Bangladesh due to vegetable consumption.

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