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Health Risk Assessment for Population via Consumption of Vegetables Grown in Soils Artificially Contaminated with Arsenic

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Authors' contributions

This work was carried out in collaboration with all authors. Authors HMZ and QFQ designed the study, managed the literatures and wrote the manuscript. Authors MIJA and RH performed the pot experiment, data recording and statistical analysis. Authors TRC, SBQ and MZIM managed As analysis of the study. All authors read and approved the final manuscript.

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ABSTRACT

Aims: A pot experiment was conducted to assess potential health risk for adult male and female human through consumption of spinach and tomato grown in soils artificially contaminated with arsenic (As).
Study Design: The experiment was laid out followed by completely randomized design (CRD) with four replications.
Place and Duration of Study: Net house of the Department of Agricultural Chemistry, Bangladesh Agricultural University, Mymensingh-2202 during the period from October 2015 to March 2016.

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Methodology: There were two types of soils (farm and industrial contaminated) and three treatment combinations viz. 0, 15 and 30 mg Kg^{-1} As for the test crops. Both spinach and tomato fruits were harvested at maturity. The amounts of As present in the dried edible and non-edible parts of the vegetables were extracted using di-acid mixture and concentrations in aqueous extracts were measured by hydride generation atomic absorption spectrophotometer (HG-AAS). To measure health risk, target hazard quotients (THQ) was calculated using the general formula established by the US EPA.
Results: Among the growth parameters length of shoot, fresh and dry weight of spinach leaves and number of leaves per plant for both spinach and tomato were comparatively higher in different treatments of contaminated soils. But average length, fresh and dry weight of roots were higher for spinach grown in farm soil. The gross yield of spinach ranged from 42.9-48.9 and 101.1-132.6 g pot^{-1} in farm and contaminated soils, respectively. The accumulation patterns of As in different parts of spinach and tomato plants were in the order- root > leaf and root > leaf > shoot > fruit, respectively. Daily As intake from spinach was almost same for both soils at treatment As-15 but it was higher for farm soil at treatment As-30. But daily As intake from tomato fruits was higher for industrial contaminated soil. Target hazard quotient (THQ) values for As was almost same due to consumption of spinach for both soils at treatment As-15 but it increased proportionally at treatment As-30 in farm soil. On the other hand, THQ values were higher for consumption of tomatoes grown in industrial contaminated soils.
Conclusion: The study results inferred that 15 mg Kg^{-1} available As in soil is enough to create health risk for population and leafy vegetables i.e. spinach is more harmful than fruit vegetables i.e. tomato. Furthermore, industrial contaminated sites are more susceptible than agricultural farm sites, and regarding health risk female are more vulnerable than male.

Keywords: Health risk; spinach; tomato; arsenic; contaminated soil.

1. INTRODUCTION

Contamination of foods by Arsenic (As) has become a challenge for both producers and consumers in Bangladesh. Soil and water pollution contribute to presence of As in foods. The occurrence of different toxic metals including As in the ecosystem is associated with rapid industrial growth, urbanization, overuse of agro-chemicals, discharge of untreated waste, sludge applications, irrigation with contaminated water, vehicular exhaust or other anthropogenic activities [1-7]. Crops and vegetables grown in As contaminated soils can be a source of As for human beings [8,9]. It has also been reported that concentrations of As in different vegetables from industrialized areas were higher than those in non-industrialized areas in various parts of the world [10]. Vegetables can take up and accumulate As in quantities high enough to cause clinical problems to humans [11]. A number of serious health problems can develop as a result of excessive uptake of As. The consumption of As-contaminated food can seriously deplete essential nutrients in the body causing, or contributing to a number of diseases. Increased frequency of spontaneous abortions and congenital malformations has been linked to arsenic exposure. In humans, chronic arsenic ingestion may cause cancers of the bladder, kidney, liver, lung, prostate, and skin [12].

Arsenic contamination of vegetables cannot be underestimated as these foodstuffs are important components of human diet. Vegetables are rich sources of vitamins, minerals, dietary fibers and fiber intake, and also have beneficial antioxidative and anti-inflammatory effects [13]. An increased metal uptake by food crops, vegetables and fruits grown on contaminated soils is often observed worldwide. Vegetables cultivated in waste water irrigated or contaminated soils acquire heavy metals in huge quantities to cause potential health risks to the consumers [14]. To assess the health risks, it is therefore important to identify the potential of a source of risk agents that may introduce into the environment, to determine the amount of risk agents that come into contact with the living environment boundaries, and evaluate the health consequence of the exposure [15].

The uptake of toxic metals in cereals and vegetables is likely to be higher and accumulation of these toxic metals in human body has created growing concern in the recent days. The daily vegetable consumption by an adult of Bangladesh is 130 g [16]. Different kinds of vegetables are grown during the year in tropical Bangladesh, but very little is known about the metal contents of vegetables [11]. Sporadic information regarding the accumulation of heavy metals in vegetables grown in industrially polluted areas in Bangladesh is available [17]. In Bangladesh, extensive As contamination of groundwater is common and the use of such water for irrigation leaving soil increasingly As rich. Vegetables grown in such As contaminated soils and consumption of those products may adversely affect human health. Considering the fact, this study was planned to assess the health risk of population through consumption of vegetables grown in arsenic contaminated soils of Bangladesh.

2. MATERIALS AND METHODS

2.1 Experimental Site

The pot experiment was carried out at the Net House, Department of Agricultural Chemistry, Faculty of Agriculture, Bangladesh Agricultural University (BAU), Mymensingh-2202, Bangladesh during the period from October 2015 to March 2016.

2.2 Collection of Soils for Experiment

Two types of soil (farm soil and industrial contaminated soil) were used for the pot experiment. Among these, farm soil was collected from the field of Genetics and Plant Breeding Farm of BAU, Mymensingh-2202, Bangladesh. The pH, EC and organic matter content of the soil were 7.18, 150 µS cm^{-1} and 1.31%, respectively. Industrial contaminated soil was collected from the site near to Noman Composite Textile Ltd., Habirbari, Bhaluka of Mymensingh. The pH, EC and organic matter content of the soil were 7.05, 312 µS cm^{-1} and 1.18%, respectively. Requisite amount of both the soils were brought to the Department of Agricultural Chemistry, BAU, Mymensingh and processed for pot experiment.

2.3 Treatments Used for the Experiment

After collection, both soil samples were analyzed for available fraction of As as described by Tessier et al. [18] and analytical results showed trace amount of available As in both soils. Due to this reason both types of soil were treated with the following treatment combinations and sodium arsenite was used as source of arsenic because aresenate uptake was strongly suppressed in the presence of phosphate, whereas arsenite transport by different uptake systems was not affected by phosphate [19,20]. The treatment combinations were: As-0 (control); As-15 (15 mg As in 1.0 Kg soil) and As-30 (30 mg As in 1.0 Kg soil). The experiment was laid out followed by completely randomized design (CRD) with four replications and thus total numbers of pots were 48 (2×3×4×2).

2.4 Test Crops and Intercultural Operations

The experiment was conducted with the seeds of spinach (*Spinacia oleraceae*) var. *Copipalong* produced by the Bangladesh Agricultural Development Corporation and seedlings of tomato (*Lycopersicon esculentum*) var. *Udayon* collected from Horticultural Farm of BAU, Mymensingh-2202, Bangladesh. Seeds of spinach were sown and seedlings of tomato were transplanted on November 12, 2015.

Fertilizers applied in the pots were as recommended for high yield goal and medium soil fertility status as described in Fertilizer Recommendation Guide [21]. The recommended doses of nitrogen (urea), phosphorus (TSP) and potassium (MoP) for spinach and tomato were 26, 8 and 8 Kg ha^{-1}, and 60, 19 and 38 Kg ha^{-1}, respectively. No manure was used in the experiment. Intercultural operations viz. weeding, irrigation, disease and pest management were done using traditional methods as and when necessary. No pesticide was applied in the experiments and distilled water was used for irrigating vegetables.

2.5 Harvesting and Processing of Samples

Spinach was harvested on January 03, 2016. Tomato fruits were harvested during early ripening stage when they attained red colour. Harvesting was started on March 14 and completed by March 28, 2016. The plant samples were tagged and taken to the laboratory where physical parameters were taken immediately and air dried for four days followed by oven drying for 72 hours until moisture content attain at desirable state. Then dried samples were ground and stored at room temperature for chemical analyses.

2.6 Chemical Analysis of Plant Samples

Powdered samples of different parts of spinach and tomato were used to prepare aqueous extract by wet oxidation method using di-acid mixture as described by Singh et al. [22]. In this method, exactly 1.00 g of finely ground plant samples were taken into a 250 mL conical flask and 10 mL of di-acid mixture (conc. HNO_3: $HClO_4$ = 2:1) was added to it. Then the flask was placed on an electric hot plate for heating at 180-200ºC temperature until the solid particles disappeared and white fumes were evolved from the flask. Then, it was cooled at room temperature, washed with distilled water and filtered into 100 mL volumetric flasks through filter paper (Whatman No. 1). Finally, the volume was made up to the mark with distilled water.

These extracts were sent to the Bangladesh Atomic Energy Commission, Dhaka and used for the determination of As content. The concentrations of As in aqueous extracts were measured by hydride generation atomic absorption spectrophotometer (HG-AAS) at 193.7 nm wavelength.

3. RESULTS AND DISCUSSION

3.1 Effect of As on Growth Parameters of Spinach

Effect of different levels of As on the number of leaves, length of leaves, fresh and dry weight of leaves of spinach were found statistically significant (Table 1). Number of leaves of spinach increased constantly in industrial contaminated soil and decreased in farm soil. The highest average number of leaves (9.38), length of leaves (19.57 cm), fresh and dry weight of leaves (12.32 and 0.93 g, respectively) were observed in spinach grown in industrial contaminated soil, while the lowest number of leaves, length of leaves, fresh and dry weight of leaves (6.97, 8.90 cm, 4.29 g and 0.41 g, respectively) were found in spinach grown in farm soil. The gross yield of spinach ranged from 42.9-48.9 and 101.1-132.6 g pot^{-1} in farm and contaminated soils, respectively. The study results inferred that all these growth parameters of spinach were increased in industrial contaminated soils, and it might be due to presence of optimum level of other essential nutrient element in that soils. A number of heavy metals at high concentrations have been reported to inhibit the vegetative growth and decrease in the productivity of crops [23]. Khan et al. [24] stated that the highest dose of Pb and Zn decreased shoot fresh weight 44 and 40%, respectively compared to control. The highest level of As (As-30) also showed similar results for most of the growth parameters of spinach.

The organ sensitivity in presence of heavy metal was in the order- root > stem > leaf as reported by Chetan and Ami [23]. May be due to that reason, length of root decreases in industrial contaminated soil than farm soil. There was a significant difference at 5% level of probability on the length, fresh and dry weight of spinach root grown in farm and industrial contaminated soils due to application of different level of As (Table 1). The highest length of root (18.82 cm) was observed in spinach grown in farm soil which

Table 1. Effect of As on different growth parameter of spinach grown in both farm and industrial contaminated soils

Treatment	No. of leaves	Length of leaves (cm)	Fresh wt. of leaves (g)	Dry wt. of leaves (g)	Moisture in leaves (%)	Length of roots (cm)	Fresh wt. of roots (g)	Dry wt. of roots (g)
Farm soil								
As-0	7.01	8.90	4.29	0.54	87.41	14.698	0.990	0.093
As-15	6.97	12.27	4.84	0.46	90.49	16.143	1.163	0.150
As-30	7.28	12.50	4.87	0.41	91.58	18.820	1.160	0.146
Mean	**7.09**	**11.22**	**4.67**	**0.47**	**89.83**	**16.550**	**1.104**	**0.130**
SD	0.17	2.02	0.33	0.07	2.16	2.090	0.099	0.030
Industrial contaminated soil								
As-0	8.52	19.57	10.10	0.83	91.78	10.493	0.960	0.092
As-15	9.38	19.16	12.31	0.93	92.45	11.005	1.095	0.114
As-30	8.87	19.56	12.32	0.75	93.91	9.693	0.860	0.079
Mean	**8.92**	**19.43**	**11.58**	**0.84**	**92.71**	**10.400**	**0.970**	**0.095**
SD	0.43	0.23	1.28	0.09	1.09	0.660	0.120	0.020
Level of significance	*	*	**	*	*	*	*	*

** = Significant at 5% level of probability; ** = Significant at 1% level of probability* was treated with As-30 and the lowest length of root (9.69 cm) was found in spinach grown in industrial contaminated soil with the same treatment. The root growth was more sensitive to the toxicity end point than shoot growth [23]. That may be a reason of lower length of spinach root grown in industrial contaminated soil compared to farm soil. Similarly, the highest fresh and dry weight for roots (1.163 and 0.146 g, respectively) were observed in spinach grown in farm soil. On the other hand, the lowest fresh and dry weight of roots (0.860 and 0.079 g, respectively) were found in spinach grown in industrial contaminated soil with control treatment. Similar result was found by Khan et al. [24], who stated that the highest dose of Pb and Zn decreased root dry weight 35 and 32%, respectively as compared to control.

3.2 Effect of As on Growth Parameters of Tomato

Effect of different levels of As on plant height, number of leaves plant^{-1}, diameter of fruits and fresh and dry weight of fruits plant^{-1} were found statistically significant at 5% level of probability (Table 2). The highest average fresh weight of fruits (55.68 g) was observed in tomato grown in farm soil which was treated with As-15. The lowest average fresh weight of fruits (26.73 g) was found in tomato grown in industrial contaminated soil with control treatment. The highest average dry weight of fruits (13.73 g) was observed in tomato grown in farm soil treated with As-30. The lowest average fresh weight of leaves (5.35 g) was found in tomato grown in industrial contaminated soil treated with As-15. Regarding fruits diameter, the maximum diameter (13.46 cm) was recorded in farm soil with As-15 treatment and the minimum (11.81 cm) was obtained from the industrial contaminated soil with the same treatment. According to Andal [25] the concentration of copper in the different parts of the plants both at high and low level of contamination was found to decrease fruit yield. There are several reports that industrial contaminated soils and sediments in Bangladesh contained higher amount of different heavy metals including Cu [26-30], which might be the reason of lower fresh and dry weight of tomato fruits grown in industrial contaminated soil.

The highest plant height (67.25 cm) of tomato was also obtained in farm soil treated with As-30 and the lowest plant height (53.33 cm) was obtained in industrial contaminated soil treated with control treatment. But the number of leaves of tomato was the lowest (68.66) in farm soil treated with no arsenic and in case of industrial contaminated soil it was almost twice (Table 2). This might be due to presence of higher amount of essential nutrient elements, which may contribute to higher vegetative growth of tomato plant and hence number of leaves were almost double in industrial contaminated soil. However, the highest number of leaves of tomato (164.50) was recorded in industrial contaminated soil treated with As-15. On the other hand, effect of different levels of As on the number of fruits plant^{-1} was found statistically insignificant (Table 2).

3.3 Concentration of As in Spinach

The highest As content (69.71 mg Kg^{-1}) in spinach leaves was found in the sample grown in farm soil which was treated with As-30 and the lowest concentration of As (trace) was recorded in plant sample grown in control treatment (Fig. 1). The maximum concentration of As in spinach root sample was 332.00 mg Kg^{-1}, which was also obtained from farm soil with the treatment of As-30. It is apparent from Fig. 1 that spinach roots are more sensitive to accumulate As than leaves and this statement is corroborate with the findings of Chetan and Ami [23]. It is also evident from Fig. 1 that relatively lower amount of As was accumulated in the spinach roots grown in industrial contaminated soil, which might be due to competition with other heavy metals present in industrial contaminated soils. However, the concentrations of As in both leaves and roots of spinach were found much higher compared with WHO maximum limit (0.1 mg Kg^{-1}), while Oti Wilberforce and Nwabue [31] found As in bitter leaf and garden egg leaf which also exceeded that limit.

3.4 Concentration of As in Tomato

Present study results revealed that the accumulation pattern of As in different parts of tomato plant was in the order- root > leaf > shoot > fruits (Fig. 2). The highest concentrations of As in tomato roots, leaves, shoots and fruits were 48.02, 5.71, 4.01 and 1.91 mg kg^{-1}, respectively for farm soil with the treatment of As-30 and 18.16, 4.19, 2.87 and 3.98 mg kg^{-1}, respectively for industrial contaminated soil with the same treatment. On the other hand, the lowest contents of As in all parts of tomato plants were obtained from the control treatment for both soils (Fig. 2). Similar to spinach, it is apparent from the study that tomato roots are more sensitive to accumulate As than other parts of the plant. This finding is at par with work reported by Chetan and Ami [23]. It is evident from Fig. 2 that relatively higher amount of As was accumulated in the edible parts of tomato (fruits) grown in industrial contaminated soil, which may create health hazard to the people. However, the concentrations of As in fruits of tomato were found hundreds-fold higher compared with WHO maximum limit (0.1 mg Kg^{-1}) for both the treatments As-15 and As-30. Present study results is corroborate with the findings of Ramteke et al. [32], and according to their report As concentrations in tomato, brinjal and amaranthus were as 0.56±0.06, 0.78±0.09 and 2.08±0.22 mg kg^{-1}, respectively.

Table 2. Effect of As on different growth parameter of tomato grown in both farm and industrial contaminated soils

Treatment	Average no. of fruits $plant^{-1}$	Average fresh wt. of fruits $plant^{-1}$ (g)	Average dry wt. of fruits $plant^{-1}$ (g)	Average diameter of fruits (cm)	Average no. of leaves $Plant^{-1}$	Average plant height (cm)
Farm soil						
As-0	11.75	43.66	11.26	12.19	68.66	57.66
As-15	6.75	55.68	12.84	13.46	81.50	55.00
As-30	5.75	48.00	13.73	13.24	103.00	67.25
Mean	8.08	49.11	12.61	12.96	84.38	59.97
SD	3.21	6.08	1.25	0.67	17.35	6.44
Industrial contaminated soil						
As-0	10.33	26.73	6.95	11.81	142.00	53.33
As-15	8.66	39.92	5.35	11.81	164.50	57.50
As-30	8.00	40.26	10.84	12.49	149.00	65.00
Mean	8.99	35.63	7.71	12.03	151.83	58.61
SD	1.20	7.71	2.82	0.39	11.51	5.91
Level of significance	ns	*	*	*	*	*

** = Significant at 5% level of probability; ns = not significant*

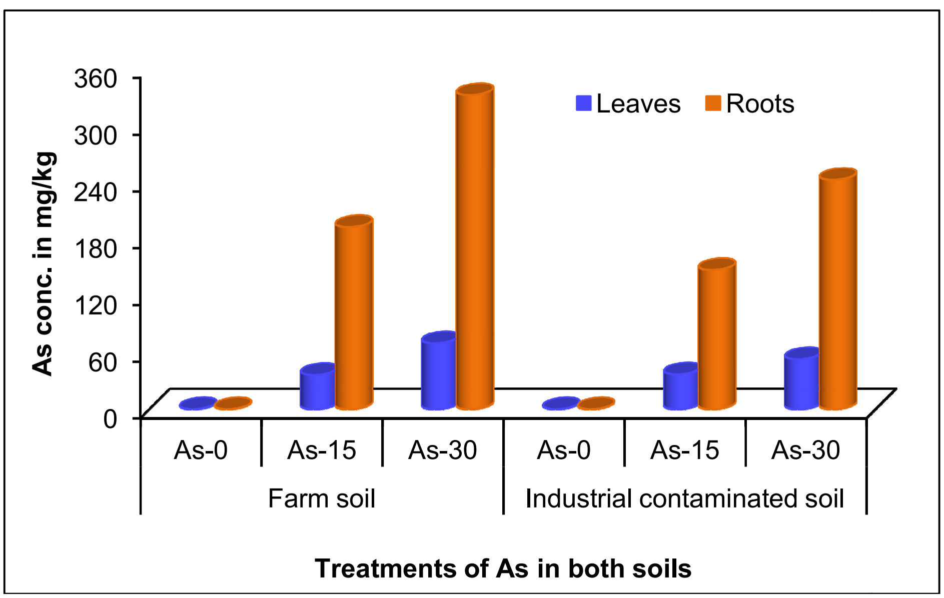

Fig. 1. Arsenic concentration (mg Kg^{-1}) in edible and non-edible part of spinach grown in both farm and industrial contaminated soils

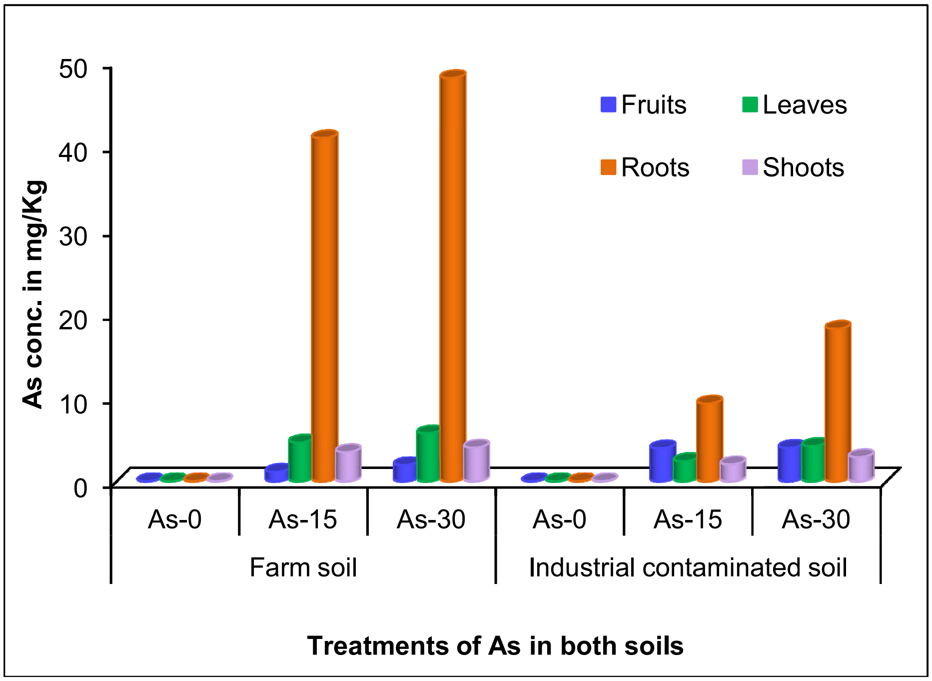

Fig. 2. Arsenic concentration (mg Kg^{-1}) in edible and non-edible part of tomato grown in both farm and industrial contaminated soils

3.5 Estimation of Daily Metal Intakes (DMI)

To appraise the health risk associated with As contamination in edible parts of spinach and tomato, the daily intake of As was calculated with the following formula-

$$DMI = (VIR \times C)/ BW$$

Where, VIR is the vegetable ingestion rate (mg $person^{-1}$ day^{-1}), C is the As concentration in edible parts of spinach and tomato samples (mg kg^{-1}, fresh weight), BW is the body weight assuming 70 Kg for adult male and 50 Kg for adult female in the present study [33].

There are several possible pathways of exposure of As to humans, but the food chain is the most important. The daily intake of As was calculated according to the average vegetable consumption for both adults male and female. A survey was conducted in March 2016 by using a prepared questionnaire of 30 family heads at industrial contaminated sites of Habirbari area of Bhaluka upazila and 50 family heads at Sutiakhali area of Mymensingh sadar upazila. Thus a total of 80 families faced the interview and in total 270 persons were effectively interviewed from two study areas. This survey data was used to calculate an average consumption rate of spinach and tomato per person per day. The survey results revealed that 10.0 and 17.0 mg of spinach and tomato as typical serving for a day for male, and 8.0 and 15.0 mg for female, respectively. The daily As intakes from spinach and tomato were calculated by multiplying the daily intake (from survey results) by the As concentrations determined in this study.

Daily intake of As from edible parts of spinach and tomato are presented in Figs. 3 and 4, respectively. It is apparent from Fig. 3 that the DMIs of As were almost same for both the soils at the treatment As-15, but the calculated values were higher at the treatment As-30 for farm soil. On the other hand, the calculated DMI values for tomato were higher in industrial contaminated soil than farm soil at both the treatments of As-15 and As-30 (Fig. 4). It is also evident from Fig. 4 that the DMI values in industrial contaminated soil were almost same for both the treatments of As-15 and As-30, which might be due to presence of other heavy metals in that soil. However, the calculated DMI values for both vegetables of the treatments As-15 and As-30 in both soils exceeded the oral reference dose for As as reported by EPA (0.0003 mg Kg^{-1} day^{-1}) [34].

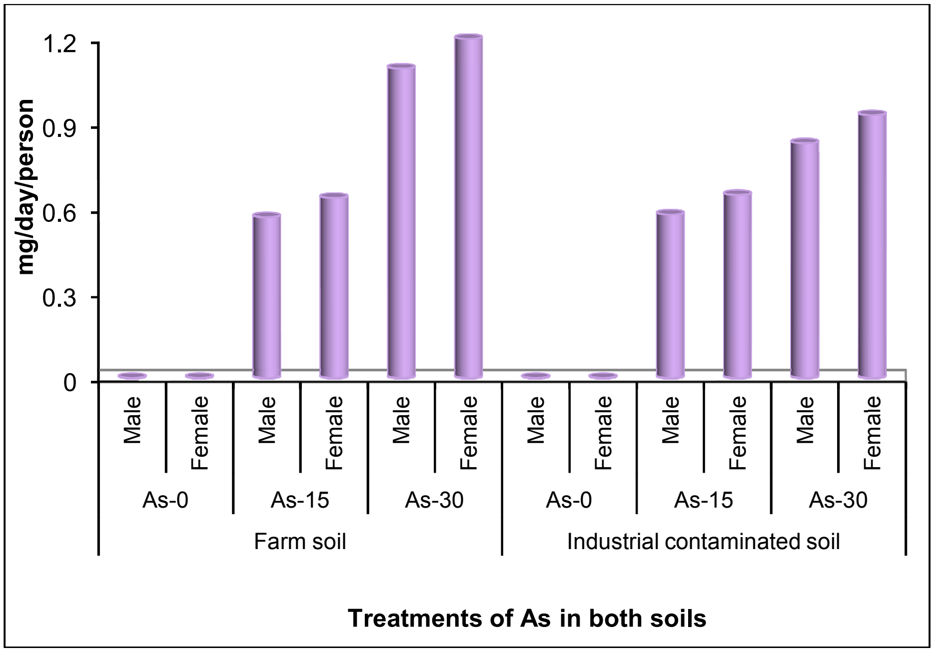

Fig. 3. Daily intake of arsenic from edible part of spinach for both male and female at different soils and treatments

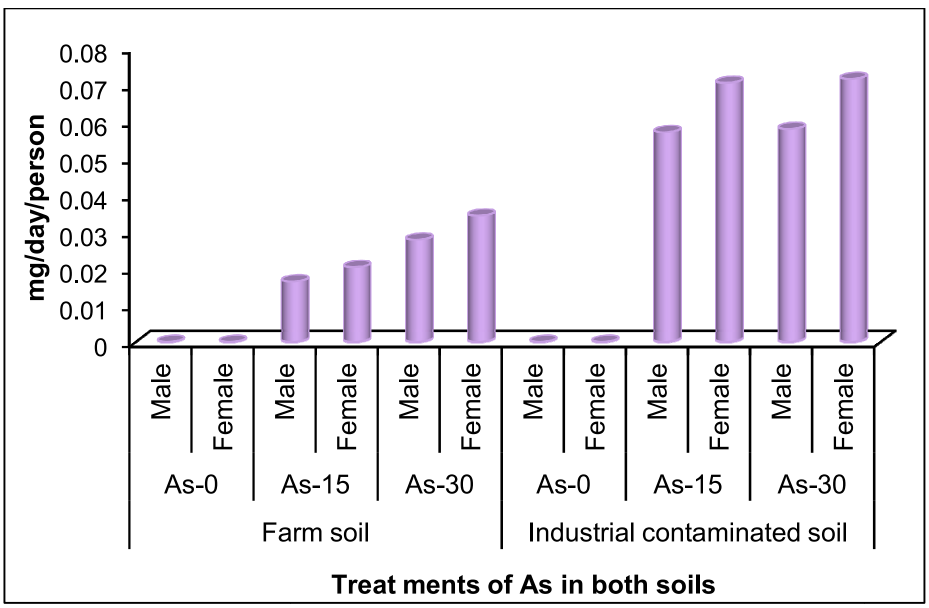

Fig. 4. Daily intake of arsenic from edible part of tomato for both male and female at different soils and treatments

3.6 Target Hazard Quotients (THQ)

The target hazard quotient (THQ) is calculated by the general formula established by the US EPA as follows-

$$THQ = (E_F \times F_D \times DMI) / (RfD \times W \times T)$$

Where, E_F is exposure frequency; F_D is the exposure duration; DIM is the daily metal ingestion (mg person^{-1} day^{-1}) and RfD is the oral reference dose (mg Kg^{-1} day^{-1}; W is the average body weight (Kg) and T is the average exposure time for noncarcinogens (365 days year^{-1} × number of exposure years).

THQ is a complex parameter that is used for the evaluation of potential health risks connected with long term exposure to toxic chemical substances [35]. The THQ <1 means the exposed population is assumed to be safe, 1 < THQ < 5 means that the exposed population is in a level of concern interval and THQ > 5 means the exposed population is in health risk. THQ parameter is a dimensionless index and THQ values are additive, but not multiplicative. It must be noted that THQ is not a measure of risk but indicates a level of concern. Target hazard quotients of As were measured considering DMI of people, average body weight (male: 70 Kg and female: 50 Kg) [33] and average life expectancy of people in Bangladesh (male: 70.6 and female: 73.1) [36].

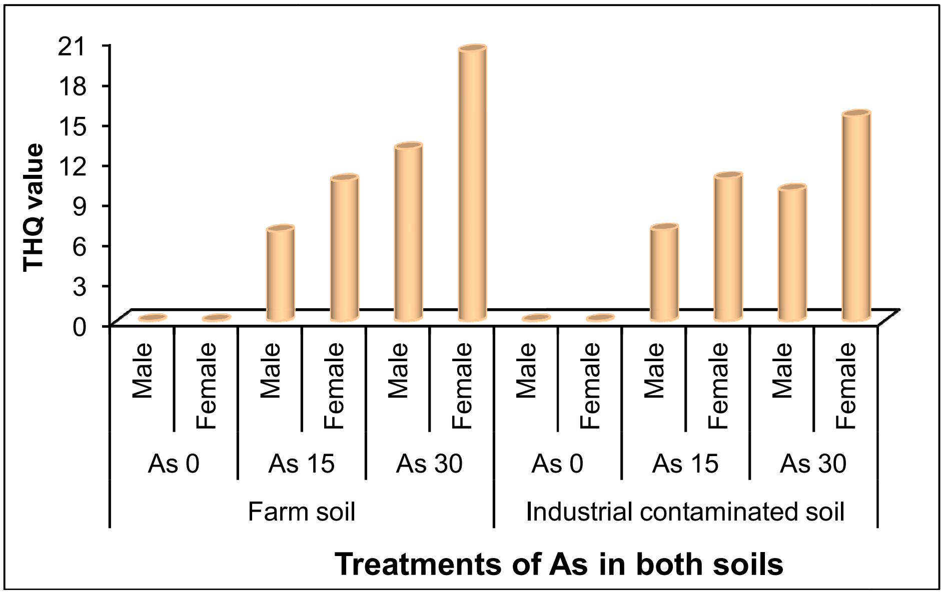

Fig. 5. Target hazard quotient (THQ) due to intake of As from spinach for both male and female at different soils and treatments

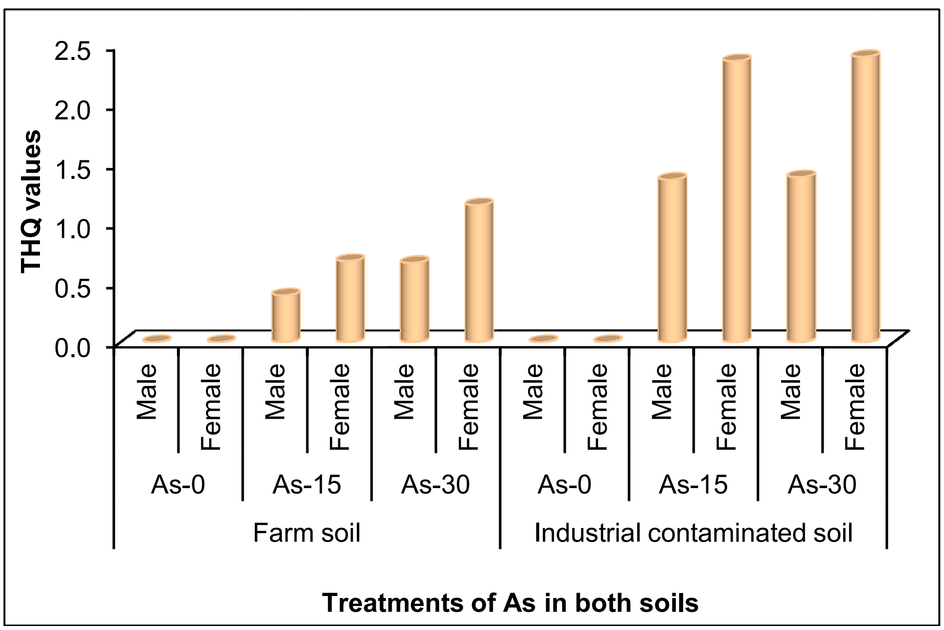

Fig. 6. Target hazard quotient (THQ) due to intake of As from tomato fruits for both male and female at different soils and treatments

THQ values due to intake of As from edible part of spinach are presented in Fig. 5 and the values varied between 6.7 to 20.2 in both soils for treatments of As-15 and As-30. So, it can be inferred from this study that the exposed population is in health risk if only 15 mg Kg^{-1} As is available in any soil and leafy vegetables like spinach is grown in that soil.

On the other hand, the calculated THQ values due to intake of As from tomato fruits are presented in Fig. 6 and the values ranged from 0.39 to 2.40 in both soils for treatments of As-15 and As-30. So, as regards to fruit vegetables like tomato, the exposed population is in a level of concern interval if 15 mg Kg^{-1} As is available in heavy metals contaminated soils. It is also apparent from both Figs. 5 and 6 that female are more vulnerable than male in relation to health risk of population in Bangladesh.

4. CONCLUSION

Extensive As contamination of groundwater in Bangladesh is common and use of such water for irrigation leaving soil increasingly As rich. Vegetables grown in such As contaminated soils and consumption of those products may adversely affect human health. The present study has revealed that the accumulation patterns of As in different parts of spinach and tomato plants were in the order- root > leaf and root > leaf > shoot > fruit, respectively. The concentrations of As in both vegetables were found hundreds-fold higher compared with WHO maximum limit for both the treatments As-15 and As-30. The study has also reported that the exposed population is in health risk if 15 mg Kg^{-1} As is available in any soil and leafy vegetables like spinach is more harmful than fruit vegetables like tomato. Furthermore, industrial contaminated sites are more susceptible than agricultural farm sites, and female are more vulnerable than male. Finally, further research at field level in arsenic contaminated soils is recommended for more consistent conclusion.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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