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Effect of Arsenic Concentration in Irrigation Water and Soil on the Arsenic Content of Vegetables in Bangladesh

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Authors' contributions

This work was carried out in collaboration among the all authors. Authors JCJ, SMIH and SK designed the study, wrote the protocol, performed the statistical analysis and wrote the first draft of the manuscript. Authors SMR, AR and MI actively involved when the experiments were conducted in Bangladesh and managed the literature searches. All authors read and approved the final manuscript.

Original Research Article

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ABSTRACT

Two pot experiments were conducted to examine the effect of arsenic (As) concentration both in irrigation water and in soil on the As content in vegetables grown in a glass greenhouse. In the first experiment, spinach (*Spinacia oleracea*), green amaranth (*Amaranthus viridis*) and gima kalmi (*Ipomoea aquatica*) were grown in soil containing 10 mgAskg^{-1}, where the irrigation water contained two As levels (0.1 and 0.5 mg L^{-1}). In the second experiment, gima kalmi (*Ipomoea aquatica*) was grown in As spiked soil at different levels [10 (control), 15, 20, 30, and 50 mgAskg^{-1} soil] and with irrigation water without As contamination. The As concentration (mg kg^{-1} DW) and As accumulation (µg plant^{-1}) in the edible part of the plants increased significantly with increasing As concentrations in irrigation water and/or soil. When plants were irrigated with As contaminated water, the As concentration of the edible part exceeded its maximum limit

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(0.5 mg kg^{-1}) in spinach and green amaranth at 0.5 mg L^{-1} of As, but gima kalmi had a smaller amount than the other vegetables. Gima kalmi had the characteristics of a lower As accumulation. Therefore, the risk level of As in irrigation water was suggested to be 0.1 mg L^{-1} for vegetables. When gima kalmi was grown in elevated levels of As contaminated soil, the As concentration of gima kalmi, even being a low As accumulator, exceeded the maximum limit at the level above 20 mgAskg^{-1} soil. The risk level of As in soil, therefore, was suggested to be 20 mg kg^{-1}. The risk value of As concentration in irrigation water and/or in soil needs to be investigated in detail by using many vegetables and/or soils.

Keywords: Arsenic; irrigation water; soil; contamination; vegetables; accumulation.

1. INTRODUCTION

Groundwater is the main source of drinking water in most of the countries including Bangladesh. It is estimated that approximately one third of the world's population uses groundwater for drinking purposes [1]. Bangladesh is a developing country in which 90% of the people depend on groundwater for drinking purposes because much of the surface water of Bangladesh is microbially unsafe to drink [2]. Though more than 80% of the population depends on agriculture for their livelihood, a lack of water during the dry season and spells of drought at the beginning and end of the rainy season is a threat to agricultural production in Bangladesh. During the last three decades, many hectares (ha) of land have been irrigated in the dry season by using shallow tube-wells (STWs). Approximately, 95% of all groundwater extracted is used for irrigation, mainly for dry season cultivation [3]. The total area under irrigation is 4 million ha and 75 percent is covered by groundwater resources: 2.4 million ha via 924,000 STWs and 0.6 million ha via 23,000 deep tube-wells (DTWs) [4].

Unfortunately, the vast area of Bangladesh groundwater is naturally contaminated with As concentrations ranging from <1–1,500 µg L^{-1}. This As concentrations in groundwater reach up to 2.0 mg L^{-1} [5, 6], in some areas in Bangladesh, where the WHO provisional guideline value for drinking water is only 0.01 mg L^{-1}. The national standard for drinking water in Bangladesh is 0.05 mg L^{-1}. According to the British Geological Survey [6], in tube wells from 41 of the total 64 districts in Bangladesh, 51% of the samples were above 0.01 mg L^{-1}, 35% were above 0.05 mg L^{-1}, 25% were above 0.10 mg L^{-1}, 8.4% were above 0.3 mg L^{-1}, and 0.1% were above 1.0 mg L^{-1}. Whereas, about 46% percent of the groundwater (both STWs and DTWs) in Bangladesh exceeds the World Health Organization (WHO) drinking water guideline (0.01 mg L^{-1}) and 27% exceeds the Bangladesh drinking water guideline (0.05 mg L^{-1}) [2, 7]. The exact percentage of STWs for irrigation is unknown because the spatial distribution of STWs for irrigation is different from that of STWs for drinking water [3].

Various survey and research studies reported different ranges of As concentrations in the irrigation water. Imamul Huq et al. [8] reported that the irrigation water As concentration varied from 0.14–0.55 mg L^{-1}. Another study showed that 87 percent of irrigation DTWs contained an As concentration of more than 0.05 mg L^{-1} and the average As concentration in those DTWs was 0.21 mg L^{-1} [9]. Ross et al. [10] estimated that 76 percent of the boro rice is grown in areas where STWs usually contain less than 0.05 mg L^{-1}, 17 percent in areas with 0.05–0.10 mg L^{-1}, and 7 percent in areas with more than 0.10 mg L^{-1}. Concentrations of As exceeding 1.0 mg L^{-1} in STWs were also reported from 17 districts in Bangladesh [11].

Arsenic is naturally present in soil all over the world, with a concentration that varies depending on the origin of the soil [12]. The background As concentration in soil is approximately 5 mg kg^{-1} [13]. Soil As concentrations ranging between 0.1–10 mg kg^{-1} are considered as non-contaminated soils [14]. The soil As concentration in Bangladesh is higher than this value and it varies depending on the location. The average As concentration in soil in Bangladesh is 12.3 mg kg^{-1}. Numerous studies documented different As concentration ranges in Bangladesh soil as follows: from 0.3–49 mg kg^{-1} [15]; from below the detection limit to 56.7 mg kg^{-1} [16]; from 46 mg kg^{-1} to less than 10 mg kg^{-1} in areas with low concentrations of As in the irrigation water [17]; and from 3.2–27.5 mg kg^{-1} [8]. In areas where irrigation water did not contain As, the soil As concentration varied from 0.10–2.75 mg kg^{-1} [8].

Only limited attention has been paid to the risks of the use of contaminated groundwater for irrigation. Irrigation water with high levels of As may result higher levels of As in the edible parts of crops (rice as well as vegetables) causing loss of yield and As contamination of the food chain, thus posing human health risks [8, 17–21]. A number of reports found evidence that irrigating with As contaminated water caused accumulation of As in the soil. Based on available data on drinking water obtained from STWs, it had been estimated that 0.9–1.36 million kg As $year^{-1}$ was brought onto the arable land via groundwater extraction for irrigation [3, 22]. According to Meharg and Rahman [17], if the irrigation water contains As of 0.1 mg L^{-1}, the water input would cause a yearly increase of 1.0 $mgAskg^{-1}$ soil. Considering the fact that the As concentration in irrigation water varies between 0.14 to 0.55 mg L^{-1}, Imamul Huq et al. [23] calculated the As loading in irrigated soils for a boro rice requiring 1000 mm of water per season to be between 1.36 and 5.50 $kgha^{-1}yr^{-1}$. Similarly, for winter wheat requiring 150 mm of irrigation water per season, As loading from irrigation water had been calculated to range between 0.12 and 0.82 $kgha^{-1}yr^{-1}$. They also calculated the loading for other crops that require irrigation and have come up with a calculated build-up of As in surface soil through irrigation. So, doubtless, As is entering the soil in different amounts day by day because of irrigating crops with As contaminated groundwater of different concentrations.

In spite of substantial research conducted by various researchers mainly on rice and wheat, very little research has been reported on vegetable crops grown in an As contaminated medium. In Bangladesh, vegetables are grown in winter season under irrigation and 0.27 million ha of land is used for vegetable crops [4]. Therefore, it is of the utmost importance to determine the As content in vegetables grown in a growth medium containing As. Limited research has examined the effects of the use of irrigation water and/or soil with As on vegetable crops. The present study was undertaken to find out the effect of the As concentration in various growth mediums on the As content in vegetables.

2. MATERIALS AND METHODS

2.1 Experimental Design

Two experiments were conducted; 1st experiment: Plants were grown in the soil slightly contaminated with As (10 mg kg^{-1} soil) where irrigation water was contaminated with As at two levels, and, 2nd experiment: Plants were grown in soil with elevated levels of As where irrigation water was not contaminated with As.

In the 1st experiment, three common leafy vegetable crops of Bangladesh *viz*., spinach (*Spinacia oleracea*), green amaranth (*Amaranthus viridis*), and gima kalmi (*Ipomoea aquatica*) were grown by irrigating with water containing two levels of As concentrations (0.1 and 0.5 mg L^{-1}). In the growth of gima kalmi, the application of 0 (control) and 0.5 mg L^{-1} As was conducted. The irrigation water was spiked with As dissolving sodium meta arsenite ($NaAsO_2$) into tap water. These two levels of As concentrations were selected depending on the As concentrations in the irrigation water of Bangladesh. The experiment was conducted to investigate the effect of varying the As concentration in irrigation water on the content of As concentration in the vegetables.

In the 2nd experiment, gima kalmi (*Ipomoea aquatica*) was grown in elevated levels of As contaminated soil. Though the original soil contained As (10 mg kg^{-1}), the soil As concentration was artificially varied by using As salt sodium meta arsenite ($NaAsO_2$) to the following levels: 10 (control), 15, 20, 30 and 50 mg kg^{-1} soil. This was done to examine the effect of increased soil As concentration on the As content in the plant. The soil As concentrations were selected up to 50 mg kg^{-1} of soil based on the information of As concentrations in Bangladesh soils [8, 15, 16]. This study was initiated to see the effect of soil As in plant if the soil is contaminated with As at different levels.

2.2 Soil Collection and Characteristics of Soil

Both of the experiments were carried out under glass greenhouse conditions in Bangladesh. The soil samples representing 0-15 cm depth from the surface were collected from the agricultural field close to the Khulna University campus, Bangladesh, by the composite soil sampling method as suggested by the United States Department of Agriculture [24]. The soil was analyzed according to the prescribed laboratory methods [25]. The bulk soil was air dried, broken, mixed thoroughly and sieved through a 2 mm sieve, and used as the growth medium in the experiments. The soil was a silty clay loam soil with 15.0% sand, 47.9% silt and 37.1% clay. The pH value of the soil was 7.93 and EC was 0.58 dS m^{-1}. The soil contained a total of the followings: As 10 mg kg^{-1}, P 0.05%, Fe 2.94%, Na 0.73%, K 1.33%, Ca 0.96% and Mg 1.47%.

2.3 Plant Cultivation

Plastic pots (2.5 L) with 2 kg of soil were used with four replications in each treatment. The soil was fertilized according to the calculation by following the Fertilizer Recommendation Guide [26] in Bangladesh. About 15–20 seeds of the plants were sown in the pots. The pots were arranged and the position was changed every day in a completely randomized way so that the plants got equal sun light. One week after sowing the seeds, the plants were thinned to 10 plants in each pot. Pesticide was sprayed as needed. During the whole growth period, all visible symptoms were observed and recorded. Only the edible part of the plant was harvested 35 days after sowing the seeds. The stems of the plants were cut at 1.0 cm above the soil. Leaves and stems were harvested together and considered to be the edible part of the plant. The harvested plant part was washed with deionized distilled water. The collected plant samples were air dried followed by oven drying at 70°±5°C for 48 hours. The dry weights of plant samples were measured and recorded. The dried plant samples were then ground and preserved for further analysis.

2.4 Measurement of the Elements

The soil and plant samples were digested with a mixture of concentrated nitric acid and perchloric acid [HNO_3:$HClO_4$, 2:1, (v/v)]. The digested plant and soil samples were analyzed for As and other elements using an atomic absorption spectrophotometer (AA-6200, Shimadzu, Kyoto) according to the previously published protocols [25, 27]. Reagent blanks and internal standards were used to ensure the accuracy and precision of the analyses. Contents of the elements are shown as mg kg^{-1} or % in the edible part of the plant. Accumulation of As in the edible part of the plant is shown as µg $plant^{-1}$.

2.5 Statistical Analyses

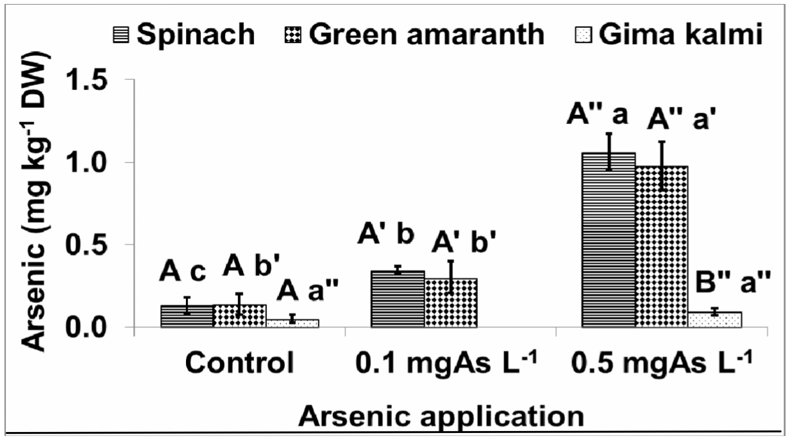
The results were expressed as the averages of four replications. The data were subjected to ANOVA. Differences between means were statistically analyzed using a Ryan-Einot-Gabriel-Welsch multiple range test (P = .05) run on the SAS software program [28] at Iwate University, Japan.

3. RESULTS

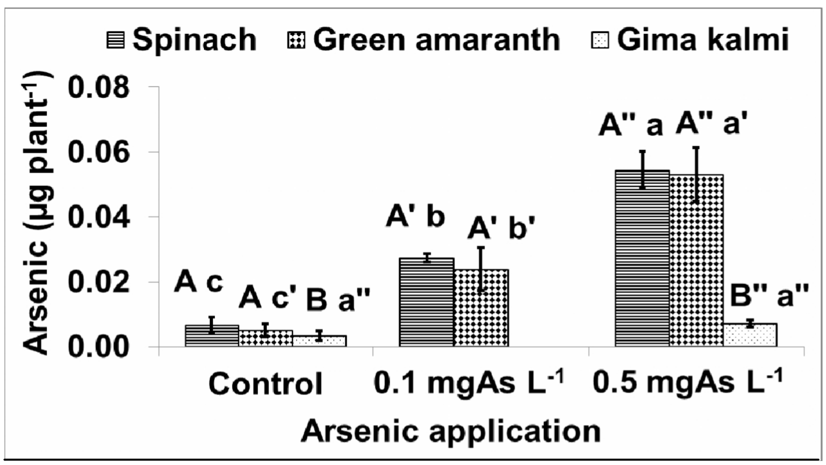
3.1 First Experiment

There were no distinct changes and no toxic symptoms among the plants. The growth of all the plants did not differ statistically, but a slightly better growth was observed in plants irrigated with water containing 0.1 $mgAsL^{-1}$. The weight of the plants irrigated with 0.5 $mgAsL^{-1}$ decreased in comparison to the plants of control and 0.1 $mgAsL^{-1}$. But it was not significantly different from any of the plant species.

The mean values of As concentration (mg kg^{-1} DW) in the edible part of the plants fed with As containing irrigation water are shown in Fig. 1a. Arsenic concentrations in the edible part of the plants increased significantly with increasing As concentrations in the irrigation water. In spinach and green amaranth, the As concentration of the edible part of the plants was significantly higher as compared with control plants, but it was not significantly different in gima kalmi (Fig. 1a). The As concentration of gima kalmi was much smaller than the other vegetables in each As application, suggesting that gima kalmi may be a low As accumulator. The As accumulation (µg $plant^{-1}$) in the edible part of the plants on the basis of dry weight is shown in Fig. 1b. Similar to the As concentration, the plants fed with As contaminated water (0.5 mg L^{-1}) accumulated a higher amount of As. Increasing the As concentration of irrigation water significantly increased the accumulation of As by the plants (Fig. 1b).



(a)

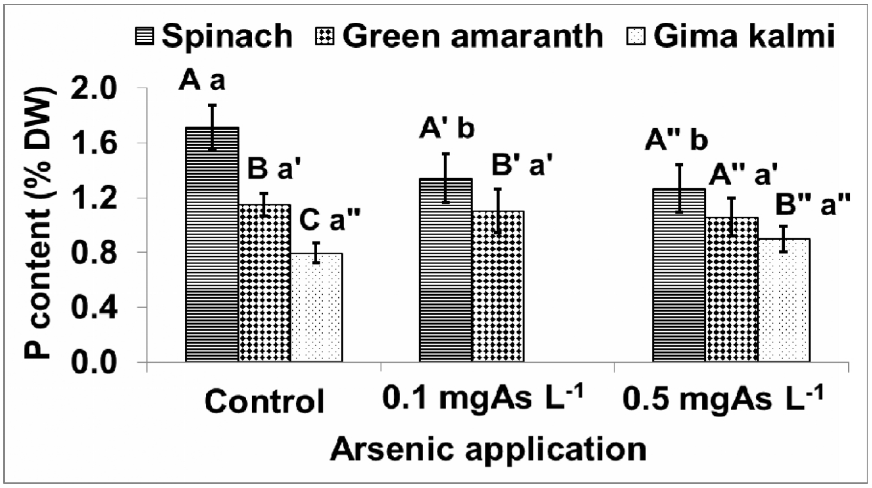


(b)

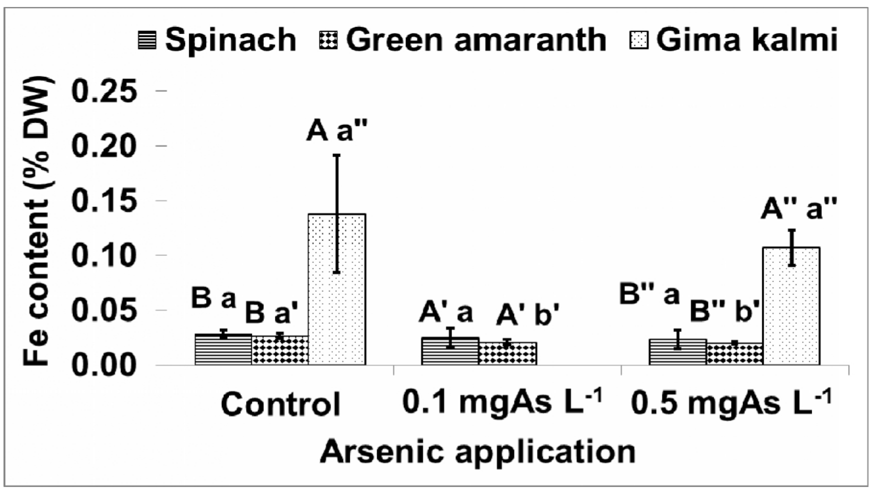
Fig. 1. (a) Arsenic concentration (mg kg^{-1} DW); and (b) Arsenic accumulation (µg $plant^{-1}$) in the edible part of the plants

Bars with different letters (small letter for As concentration among the treatments and capital letters for among the three plants) are significantly different (*P* = .05) according to a Ryan–Einot–Gabriel–Welsch multiple range test. Error bars are the standard deviations (SDs). In gima kalmi, the application of 0 (control) and 0.5 mg L^{-1} As was conducted.

The P concentration in the edible part of spinach significantly decreased by As application, but it was statistically similar in green amaranth and gima kalmi (Fig. 2a). The P concentration was generally higher in spinach than green amaranth and gima kalmi in all As applications. There was, however, no prominent effect of As application on the Fe concentration of the plant part (Fig. 2b). In green amaranth, the Fe content was slightly decreased. Gima kalmi had a higher amount of Fe than spinach and green amaranth.



(a)



(b)

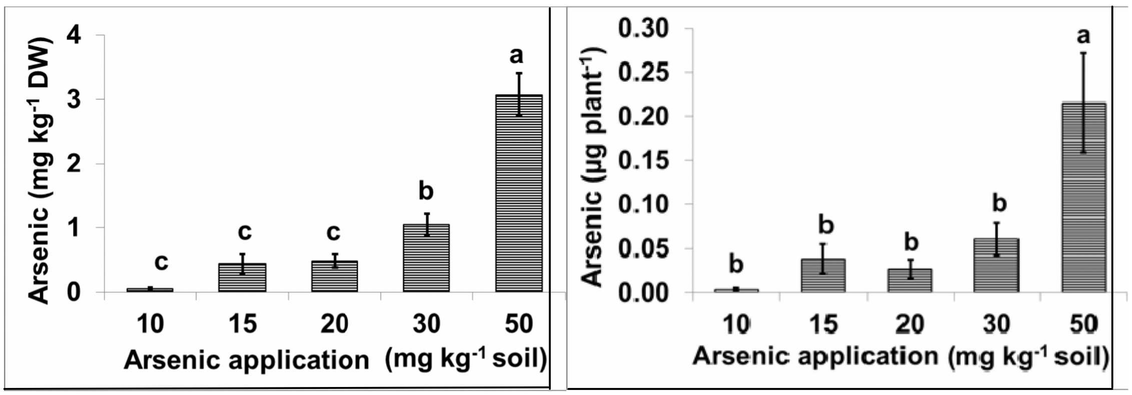
Fig. 2. (a) P content (%); and (b) Fe content (%) in the edible part of the plants

Bars with different letters (small letter for As concentration among the treatments and capital letters for among the three plants) are significantly different (*P* = .05) according to a Ryan–Einot–Gabriel–Welsch multiple range test. Error bars are the standard deviations (SDs). In gima kalmi, the application of 0 (control) and 0.5 mg L^{-1} As was conducted.

3.2 Second Experiment

This experiment was conducted only with growing gima kalmi, a low As accumulator. Similarly to the 1st experiment, no distinct changes and no toxic symptoms among the growing plants were observed during the growth period. The dry weights of all the plants were statistically similar, but a slightly better growth was observed in plants grown in soil with a lower As concentration (15 mg kg^{-1} soil) than the other plants.

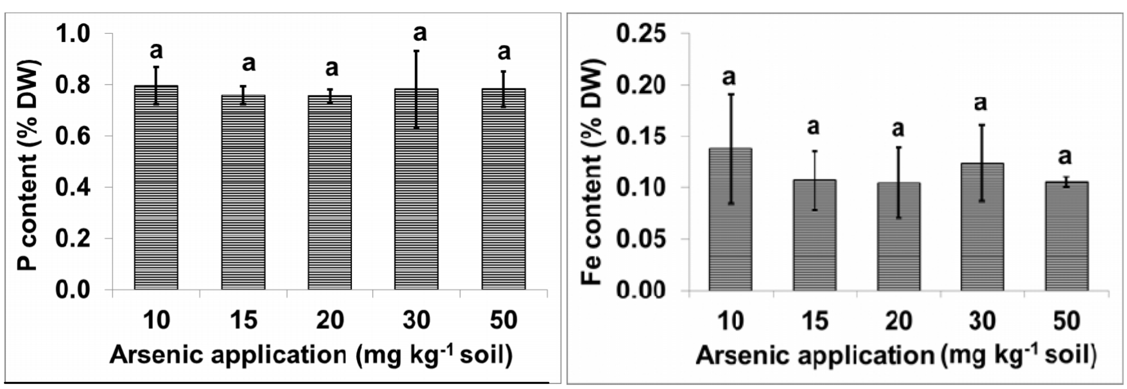
The mean values of the As concentration (mg kg^{-1} DW) and As accumulation (μg $plant^{-1}$) in the edible part of the plant gima kalmi are shown in Figs. 3a and 3b, respectively. The As accumulation (μg $plant^{-1}$) in the plant part was also calculated on the basis of the dry weights of the plants. The As concentrations in the edible part of the plant increased significantly with increasing As concentrations in the soil. The As concentration in the edible part of the plants was significantly higher in the soil with higher As concentrations (30 and 50 mgAskg^{-1} soil), but there were no significant differences in the soil with lower As concentrations (15 and 20 mgAskg^{-1} soil) (Fig. 3a). Similar to the As concentration in plants, the As accumulation in plants significantly increased when grown in soil with elevated As (50 mgAskg^{-1} soil) (Fig. 3b) and it was 70 times higher than that of control. The results also showed that there was no effect of As on the content of P and Fe in the edible part of the plants (Figs. 4a, b).



(a) (b)

Fig. 3. (a) Arsenic concentration (mg kg^{-1} DW); and (b) Arsenic accumulation (μg $plant^{-1}$) in the edible part of gima kalmi

Bars with different letters are significantly different (*P* = .05) according to a Ryan–Einot–Gabriel–Welsch multiple range test. Error bars are the standard deviations (SDs).



(a) (b)

Fig. 4. (a) Phosphorous (%); and (b) Fe (%) contents in the edible part of gima kalmi

Bars with different letters are significantly different (*P* = .05) according to a Ryan–Einot–Gabriel–Welsch multiple range test. Error bars are the standard deviations (SDs).

4. DISCUSSIONS

It was shown that a slightly better growth of the plants was observed with lower As concentrations both in irrigation water and in soil as compared to the control in both the experiments. The changes in the growth of the plants, however, were not significantly different. It was reported in a study using hydroponic culture that the growth of the plants to some extent increased at lower doses of As [29]. In contrast, a decreased growth of the plants grown in an As containing medium was also reported by other researchers [30–32].

The application of As with irrigation water and mixing in soil increased the As concentration in the edible part of the plants significantly with increasing levels of As in the soil and irrigation water. The As concentration in the edible plant part exceeded the maximum limit of As (0.5 mg kg^{-1}) for vegetables [33]. It can be noted that there is no universal limit value of As in vegetables but only China has the maximum limit value of As for vegetables (0.5 mg kg^{-1}), though some other countries (Australia, New Zealand, Singapore and UK) has the maximum limit value of As for cereals. The maximum limit value in rice in those countries is 1.0 $mgAskg^{-1}$ and that of India is 1.1 $mgAskg^{-1}$ [33]. International risk level of As in vegetables may be necessary.

In the 1st experiment, the As concentrations in the edible part of the plants increased with increasing As levels in the irrigation water. The As concentration in vegetables grown with irrigation water containing 0.1 $mgAsL^{-1}$ was less than the maximum limit for the vegetables, but it was two times higher than the maximum limit when the irrigation water contained 0.5 $mgAsL^{-1}$ in both spinach and green amaranth as shown in Figure 1a. On the other hand, the As concentration in gima kalmi was lower than the maximum limit. The concentration of As differed depending on the species of vegetables. It was clear that gima kalmi had the characteristic to have a lower As concentration in the edible part than spinach and green amaranth. The plant seemed to be a low As accumulator. The As concentration in plants followed in the order: spinach>green amaranth>gima kalmi. In this experiment, the soil contained As of 10 mg kg^{-1}. All of the plants were grown in a low level of As contaminated rhizosphere. In this condition, the As concentration of the control plants was very low. It was one-fourth of the maximum limit (0.5 $mgAskg^{-1}$) in spinach and green amaranth and one-tenth in gima kalmi. In contrast, the As concentration of the control plant of the vegetables was about one-tenth and half in gima kalmi as compared with that of the plants with irrigation water of 0.5 $mgAsL^{-1}$. Some other researchers also showed that through irrigation water there is an accumulation of As in the crops [8, 17, 20, 34]. Both organic and inorganic forms of As are present in terrestrial plants [35] but As in terrestrial food plants is dominated by inorganic As [36] and in case of peas and spinach only inorganic As was present. In our experiments, we measured the total As contents in the plant by digesting the plant samples with a mixture of HNO_3 and $HClO_4$. In this experiment, organic and inorganic As could not be separately measured. It is also reported that in the Bengal Delta region, where As-contaminated water has been used for irrigation, and that some vegetables and spices contained relatively high concentration of As [37], of which most of the As are present only in inorganic forms [37, 38].

Even if the As concentration of the soil was low (10 mg kg^{-1}), the presence of As in the irrigation water in the cultivation of the vegetables constitutes a risk for elevating the As in the plant tissues. Based on the current result, the As concentration in irrigation water up to 0.1 $mgAsL^{-1}$ would be the safe level for cultivation of the vegetables considering the maximum limit. But practically, it may be impossible in Bangladesh depending on the location.

In the 2nd experiment, the As concentration in the plant (gima kalmi) grown in different levels of As contaminated soil exceeded the maximum limit for vegetables of 0.5 $mgAskg^{-1}$ [33] when the soil As concentration was higher than 20 mg kg^{-1}. The As concentration in the edible part was two and six times higher than the maximum limit for vegetables of 0.5 $mgAskg^{-1}$ when grown in 30 and 50 $mgAskg^{-1}$ soil, respectively (Fig. 3a). In contrast, the concentrations of As in the plant grown in 15 and 20 $mgAskg^{-1}$ soils were very close (0.44 and 0.48 $mgAskg^{-1}$, respectively) to the maximum limit. For the control plants grown in 10 $mgAskg^{-1}$ soil, the As concentration in plants was very low (0.05 $mgAskg^{-1}$), which is one-tenth of the maximum limit (Fig. 3a). It is known that the concentrations of As in non-contaminated soils range from 0.1 to 10 $mgAskg^{-1}$ [14]. In our 1st experiment, the As concentration in soil was within the range of non-contaminated soil and the As concentration in the plants grown with non-contaminated water (control) was very low as compared with the maximum limit. Therefore, it was considered that the As concentration in soil having higher than 20 $mgAskg^{-1}$ increased the As concentration in plants. It seemed clear that if the soil contained As higher than 20 mg kg^{-1}, the vegetable grown in it would exceed the maximal limit even in gima kalmi, a low As accumulator. It is considered that the As concentration in the edible part of the other vegetables must exceed the maximal limit when grown in the soil having As higher than 20 mg kg^{-1}. The risk value of the soil As concentration may differ depending on the climate, soil type, and species of vegetables. Based on the result of gima kalmi, a low As accumulator, it can be noted that vegetables should not be cultivated in the soil with an As concentration higher than 20 mg kg^{-1} soil. More research needs to be conducted using many types of vegetables in other countries for the safety of food production. As shown in the 1st and 2nd experiments, the As accumulation (µg $plant^{-1}$) in plants also increased significantly with increasing As concentrations in the irrigation water or the soil. The risk of higher concentrations of As in irrigation water and/or soil in the vegetable cultivation was clearly shown.

Phosphorus is an important nutrient for energy transfer and protein metabolism in plants [39]. There was no change in the P concentration in the edible part of any of the plants (Figs. 2a, 4a). However, the concentration of P in the plant part decreased significantly only in spinach as compared with control (Fig. 2a). The P concentration in spinach, however, was higher than in green amaranth and gima kalmi in the 1st experiment irrespective of the As application through irrigation water (Fig. 2a). The tendency to reduce the P contents by the application of As in edible plant part of spinach was probably due to either As phytotoxicity or the competitive uptake of As and P [40]. Spinach may be sensitive to As in P absorption, but the P content in other plants was not affected by As absorption. It is known that PO_4 competes with arsenate on the transporter for absorption at the root surface [41, 42].

The Fe content was much higher in gima kalmi than that of spinach and green amaranth (Fig. 2b). There is a possibility that the higher Fe concentration may be related physiologically to the characteristic of low As accumulation in the edible part of the plant. It is known that As may induce Fe deficiency in plants grown hydroponically [43]. There may be a deficiency of Fe concentration depending on the plant part, but the difference in the Fe concentration was not so prominent in the current experiment. Decreasing Fe concentrations have previously been reported in plant tissues exposed to an enhanced As level [29, 44, 45]. However, increased Fe uptake under As stress condition also reported [46].

5 CONCLUSIONS

Considering the maximum limit of As in vegetables, the plants fed with irrigation water with an As concentration exceeding 0.1 mg L^{-1} might produce vegetables highly contaminated with As depending on the species. In order to reduce the risk of As toxicity in the people through consumption of the vegetables, irrigation water containing less than 0.1 $mgAsL^{-1}$ should be applied for irrigation to the vegetables. Gima kalmi was shown to be a low As accumulator when As contaminated water was applied.

The soil As value of 20 $mgAskg^{-1}$ might be a risk level for increasing the As concentration of vegetables considering the maximum limit. This result was shown by using gima kalmi, lower As accumulator than the other vegetables. For the other vegetables, the risk level for As in soil may be lower than 20 mg kg^{-1}. The value may be different depending on the soil type also. However, 20 $mgAskg^{-1}$ may be suggested as a risk level for vegetables because even gima kalmi, a low As accumulator, exceeded the maximum limit in China of As in its edible part. It is considered that if the soil As concentration is higher than 20 $mgAskg^{-1}$, vegetables should not be cultivated even though As-free irrigation water is applied. This report is one of the steps to assess the risk level of As in irrigation water and/or soil in the cultivation of vegetables. In order to determine the critical level or risk level of As concentration in irrigation water and/or in soil for individual vegetables, extensive experiments will be required.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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