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Analysis of Arsenic Concentrations and Correlation in Water, Soil and Aurum by Neutron Activation Analysis Technique: A Case Study in Bagerhat, Bangladesh

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Abstract Arsenic concentrations were measured in water, soil and arum (vegetables) samples using the Neutron Activation Analysis method and a correlation between arsenic concentrations in the samples was investigated. The case study at Bagerhat, Bangladesh revealed that almost all the water samples were contaminated by a hazardous level of arsenic that exceeding the World Health Organization recommended value of 0.05 mg/L for Bangladesh. Arsenic concentration of all the water samples ranged from 0.09 to 0.87 mg/L. The concentrations in soil and aurum samples were found to be in the range of 2.22–35.21 and 0.07–0.73 mg/kg, respectively. A positive correlation between arsenic concentrations in soil and water samples was observed. Aurum sample was found to be contaminated by arsenic to a harmful level if the corresponding water sample was also highly contaminated.

Keywords Arsenic · Neutron activation analysis · Aurum · Same origin

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A hazardous level of natural arsenic pollution of ground water has been a great threat for a vast region (especially in the Southern and South-western parts) of Bangladesh (Moinuddin 2004). About 57–80 million people are reported to be at a possible health hazard of arsenic exposure for drinking water with arsenic above the WHO (World Health Organization) provisional guideline value (>0.01 mg/L) (Dhaka Community Hospital 2010; Yu Chen et al. 2009). Several thousands of people have already been suffering from chronic arsenicosis in the affected regions (Bhattacharya et al. 2007). Arsenic contamination has now spread to soil and sediment (Meharg and Rahman 2002; Patel et al. 2005), food stuffs (Al Rmalli et al. 2004), herb plants, herbal products (Saper et al. 2004; Islam et al. 2007) and other environmental components such as chicken (Tamar Lasky et al. 2004), human nail, hair, urine (Chowdhury et al. 2000) date juice and coconut water (Correspondent 2010). The spread of arsenic contamination has been a serious health hazard to the people of contaminated regions and is creating social problems (Dhaka Community Hospital 2010) in Bangladesh.

Bangladesh mainly depends on agricultural sector for its gross domestic production (GDP), and about 86% of the total water requirements are used from groundwater (BADC 2002). The use of arsenic contaminated water for irrigation has become a serious threat to the sustainable agriculture in the affected regions and pose an additional health hazard from the irrigated crops (Norton et al. 2010). Because of irrigation with arsenic contaminated water, concentration levels in soil are also increasing over a period of time, and these levels can reach up to 57–83 μg/g arsenic (Ullah 1998; Alam and sattar 2000; Alam and Sattar 2000; Polizzotto et al. 2006; Ravenscroft 2009). A significant quantity of vegetables items (e.g., aurum) are also grown in Bangladesh by using tube-well water or hand pump water. One study found a harmful level of arsenic was present in all the vegetables exported to the United Kingdom from Bangladesh and the highest arsenic values were found to be in the skin of Arum tuber, 540 μg/kg, followed by Arum Stem, 168 μg/kg, and Amaranthus, 160 μg/kg (Al Rmalli et al. 2004). However, aurum is an iron affluent and one of the cheapest vegetables items found everywhere in Bangladesh. Pregnant women in Bangladesh consume this item to compensate the post pregnancy deficiency, but its adverse effect on pregnancy was reported due to chronic arsenic exposure (Hasnat et al. 2005). Very few researches have been done so far to test the possible mitigation measures and the people in the affected areas are yet to be aware about these new threats. These concerns signify the analysis of arsenic concentrations in water, soil and aurum (vegetable-item) and their correlation for the specific sites of sampling. The present study concentrates to one of the most arsenic contaminated districts (Bagerhat) located in the South-western part of Bangladesh with a mean arsenic concentration of 156 μg/L in groundwater (BGS 2000). About 66% of groundwater from the shallow aquifer (less than 150 m deep) of this region exceeds the Bangladesh standard for arsenic of 0.05 mg/L. In particular, this analysis was aimed to determine the magnitude of arsenic concentrations in tube-well water, soil and in an iron affluent vegetable item, aurum collected from the same origin. An attempt was also made to investigate whether there exists any correlation between arsenic concentrations in the three types of samples (water, soil and aurum).

Materials and Methods

A total of 18 sets sample were collected from three different types of sources (water, soil and aurum) across Bagerhat district of Bangladesh. To collect each set of the samples from the same origin, the sample sites were selected maintaining the same environmental conditions. Both soil and aurum samples were collected from the surrounding areas of the tube-wells selected for the ground water sampling. Tube-well users and vegetables (aurum) growers were interviewed during the collection of each set of the samples. Shallow types of tube-wells were selected for water samples as these are the ones that are commonly used in Bangladesh for drinking and irrigation purposes. The sample containing bags and bottles were marked with sample identification on their outer surfaces.

The pre-selected tube wells were pressed approximately 20 times before collecting each of the water samples. Each 100 ml water sample was collected in a polypropylene bottle and then preserved adding five milliliter of 37% hydrochloric acid. The collected samples were processed to make ready for irradiation in the reactor lab at the Atomic Energy Commission (AEC) of Bangladesh. A quantity of 400 μl solution from each of the preserved water samples was dropped on a six fold filter paper using a fine pipette. In order to measure the absorbed water, the water soaked filter paper was then weighed using a microbalance. Water soaked filter paper was dried and wrapped in a twofold polyethylene paper and was then heat sealed. In a similar way, 20 μl of 1000 mg/L and 200 μl of 4.42 mg/L IAEA arsenic standard reference water materials were prepared for irradiation.

The soil and aurum samples were collected in acetone-cleaned film containers and preserved in a cool dry place. The soil samples (surface soils) were collected from the rooting zones of the aurum plants selected for the aurum samples. Samples were then transferred to acetone-cleaned Petri dishes and dried up in an oven at 70 degree (for 40–50 h) until a constant weight was achieved.

The samples were then crushed in an agate mortar to minimize sampling inhomogeneities and then preserved with proper identification as stock samples (about 50 mg of each).

About 40 mg of IAEA-336 standard sample (Lichen) was also prepared and was poured into separate clean polyethylene bag as the reference stock sample. The stock samples were then encapsulated in polyethylene papers and the sample codes were marked on the outer surfaces of the papers. The samples, standard reference materials and blanks were divided into several sets for irradiation and counting purpose. All sets of the samples including the respective standards were then poured into the marked vials to get them ready for irradiation.

The processed samples and the respective standards were irradiated using a thermal neutron beam of flux 7.6×10^{12} n/cm^2/s in the Dry Central Thimble (DCT) facility of three-MW TRIGA Mark-II research reactor located at the Atomic Energy Research Establishment (AERE), Dhaka, Bangladesh. The emitted characteristic γ-rays from the radioactive samples were then detected by a High Purity Germanium (HPGe) detector and the energy spectra were analyzed using a computer based Canberra

Table 1 Standard deviations of the certified values of the reference materials from that of the experimental values

Std. materials	IAEA Soil-7	(As std) Lichen	(As std) Water[1]	(As std) Water[2]
Measured values (mg/L)	13.43	0.652	1005.02	4.51
Certified values (mg/L)	13.40	0.639	1000.00	4.42
(%) RSD	0.20	0.034	0.50	2.03

S-100 Multi Channel Analyzer (MCA). The radioactive samples emitting γ-ray of energy 559.1 keV was identified to be originated from arsenic indicating arsenic contamination. The resolution of the detector was determined using a calibrated ^{60}Co source and was found to be 1.93 keV at the 1332.52 keV gamma line.

In order to eliminate the effect of short lived nuclides, 2 days of cooling period were allowed for the irradiated samples before the emitted γ-rays from them were counted. Lead shield was used to minimize the background radiation coming from cosmic rays and other natural or nuclear sources. A measuring time of 1800 s was allowed for arsenic detection for each of the samples. The lowest concentration found in each set of the samples was used to determine the detection limit (DL) of that sample type. The DLs for water, soil and aurum were observed to be 0.027, 0.25 and 0.031 mg/L, respectively.

The elemental concentrations in the samples were found to be well above the detection limits. The results were found to be accurate within 0.04–2.03% of the certified values as shown in Table 1.

Results and Discussion

Figures 1, 2 and 3 represent the arsenic concentrations in water, arum and soil samples, respectively. Arsenic was detected in all water samples ranging from 0.09 to 0.87 mg/L. Arsenic was also found in all soil samples and in eleven aurum samples out of eighteen samples. The concentrations in soil and aurum samples were found to be in the range of 2.22–35.21 and 0.07–0.73 mg/kg, respectively. According to the maximum permissible limit 0.05 mg/L for Bangladesh, all water samples but one exceeded the limit. However, all water samples were contaminated by arsenic at a harmful level compared to the WHO standard for arsenic in water (0.01 mg/L). The arsenic concentrations in most water samples are several times higher than the recommended value. Therefore, it can be said that most of the tube-wells in Bagerhat with their present status are not suitable for drinking and irrigation purposes. All aurum samples were not contaminated by arsenic but it is evident that arsenic was present only when the corresponding water or soil sample was contaminated by arsenic at a harmful level. It can, therefore, be concluded that crops grown in such arsenic contaminated soil will be contaminated by arsenic as well.

The safe levels of arsenic in soil and food-stuffs for Bangladesh are not clearly defined in literature. The safety standards of arsenic in soil and food used by the developed countries might be different for Bangladesh where most people live on rice and use tube-well water for drinking purposes (Duxbury 2005). A study, carried out by Jahiruddin

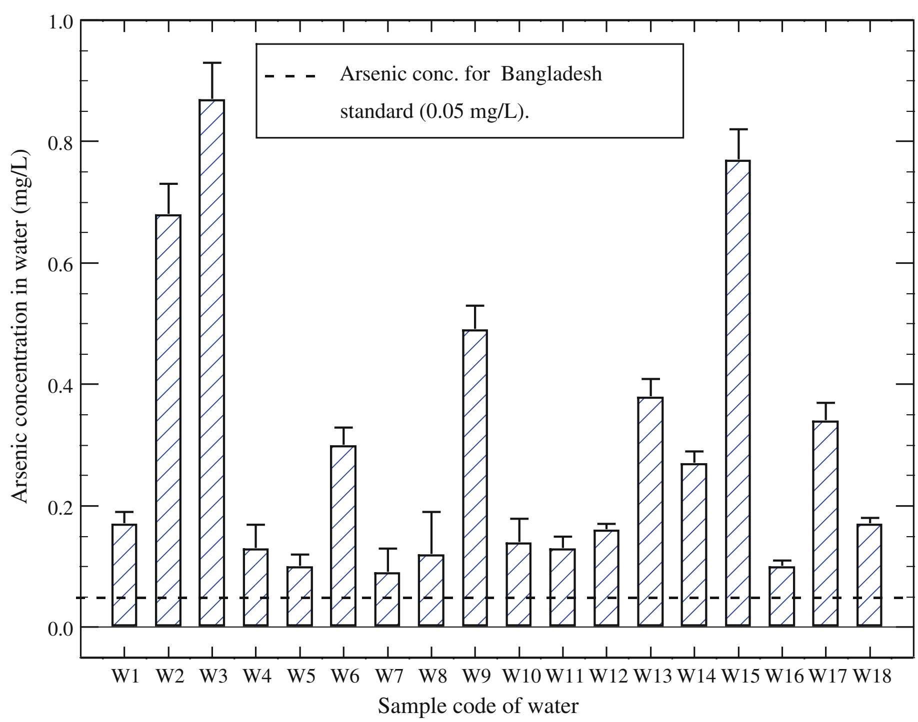

Fig. 1 Arsenic concentrations in the water samples collected from Bagerhat

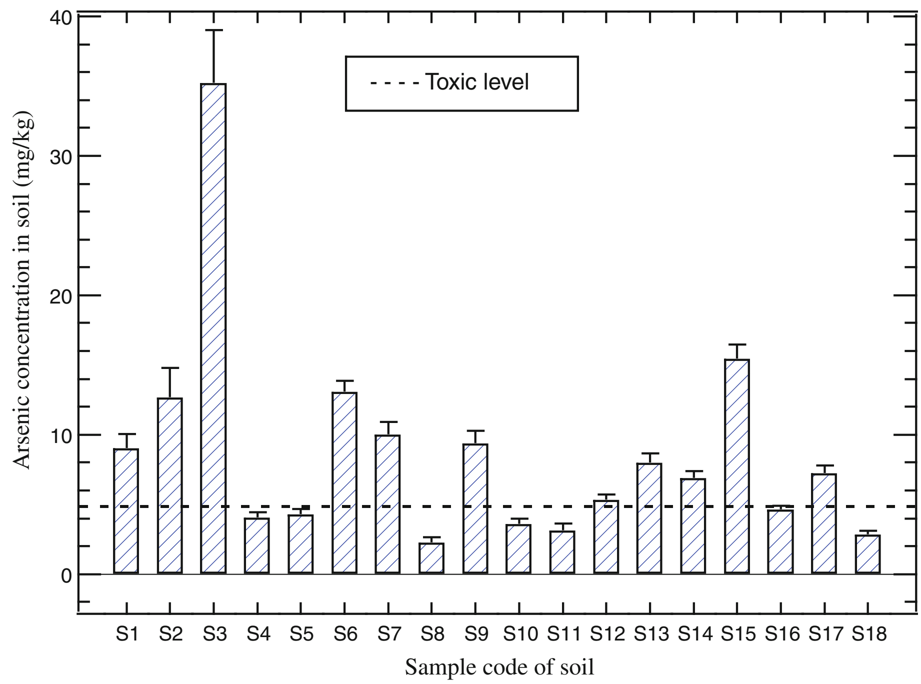

Fig. 2 Arsenic concentrations in the soil samples

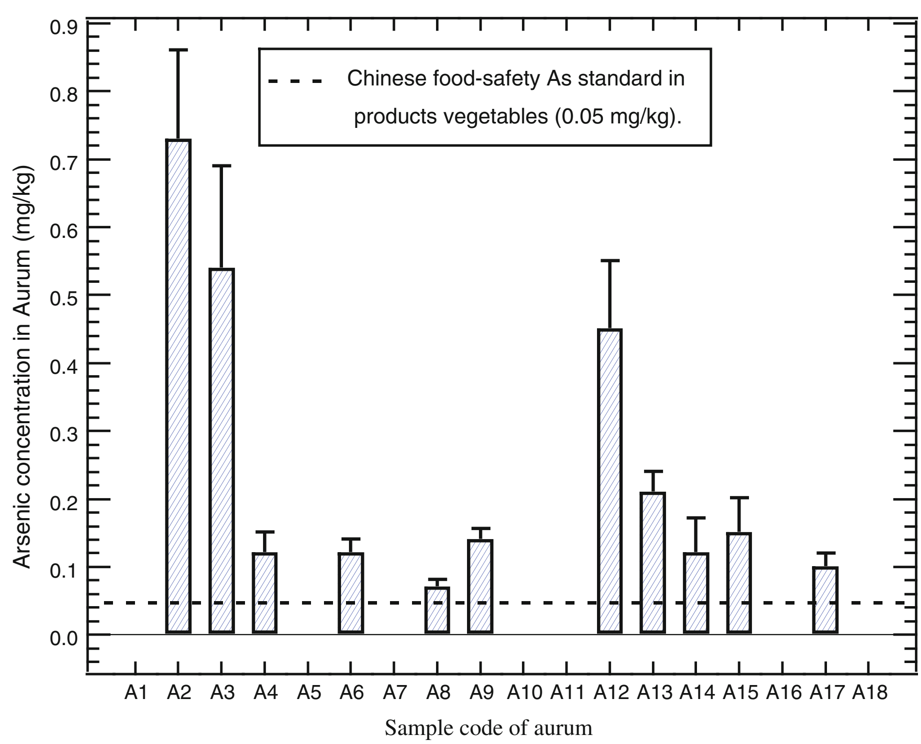

Fig. 3 Arsenic concentrations in the Aurum samples

et al. (2004) reports that the arsenic contamination of irrigation water by 1 mg/L and soil by 5 mg/kg reduces the yield of (Boro) rice (Jahiruddin et al. 2004). It is also reported that vegetables are more sensitive to arsenic contamination of soil and irrigation water. According to the Chinese food safety standard for inorganic arsenic in various vegetable products is 0.05 mg/kg (Bugang 2006).

In the present study, the safety levels for arsenic in soil and in vegetables item (aurum) were taken as 5 and 0.05 mg/kg, respectively in the context of Jahiruddin's report and Chinese food safety standard. According to these limits, ten soil samples of eighteen and eleven aurum samples of eighteen were exceeded these levels. From the results of the present experiment, Bagerhat district appears

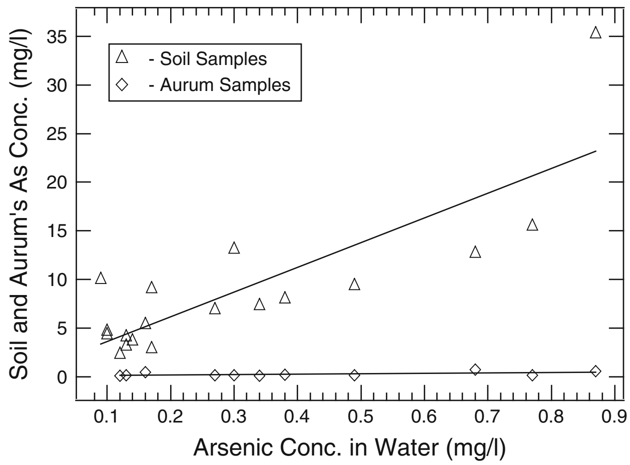

Fig. 4 Correlation of As concentrations between water and soil and Aurum Samples

to be highly arsenic contaminated region. The measurements were carried out at a reactor based laboratory using NAA method while accurate measurements of arsenic at low levels relevant to health requires laboratory analysis.

Figure 4 shows a correlation between arsenic concentrations in water and soil. A typical positive correlation was found between arsenic concentrations in water and soil samples. In Fig. 4, it is evident that arsenic contamination in soil increases with the increasing contamination in water. Contaminated soil could thus be a supportive ally of contaminated water to spread arsenic toxicities to the people living in the arsenic affected regions. No correlation was found in arsenic contamination between water and aurum.

Arsenic contamination of a (sample) plant could depend on the arsenic absorbency capacity of the plant, growing-time of the plant by arsenic contaminated water and the As-levels in the soil. The correlation study of arsenic concentrations needs these factors to be investigated precisely for more accurate result. In general, collecting water, soil and plant samples from the same origin is, very often, difficult. However, a clear understanding of arsenic transfer mechanism and correlation of arsenic concentrations in soil, water and plants is crucial for addressing the issue rigorously. An experiment could be carried out by growing an absorbent type of plant with arsenic contaminated water for a specific period of time (e.g., 1 year). A set of water, soil and plant samples could then be collected to investigate the arsenic transfer mechanism and correlation processes in the samples precisely as a function of time. Also, a time dependent positive relation between arsenic concentrations in different environmental components could be addressed precisely.

This finding could be important for arsenic detection in other food-stuffs grown in arsenic contaminated water. A long time monitoring of arsenic contamination of different environmental components under different circumstances is vital for Bangladesh. Establishment of the safe levels of arsenic standards in food-stuffs and soil for Bangladesh is also suggested considering the circumstances in Bangladesh, and in doing so the standard values used in the developed countries should be taken into account.

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