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Slow arsenic poisoning of the contaminated groundwater users

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ABSTRACT: This paper gives impact of Arsenic contaminated water on human health as well as overview of the extent and severity of groundwater arsenic contamination in Bangladesh. Scalp hair is the most important part of the human body to monitor the accumulation of this type of poison. Therefore, an experiment has been carried out by Neutron Activation Analysis (NAA) at Atomic Energy Research Establishment (AERE), Savar, Dhaka, Bangladesh on human hair of corresponding tube well water users of these areas to determine the total accumulation of arsenic to their body. Hair samples collected from the region where the groundwater was found highly contaminated with arsenic. The obtained results of arsenic concentration in the lower age (Hb) categories of users (below 12 years of age users) is in the range of 0.33 to 3.29 μg/g (ppm) and that in the Hu categories (upper 12 years of age users) is 0.47 to 6.64 μg/g (ppm). Where as maximum permissible range is 1 ppm certified from WHO. Results show that the peoples are highly affected where the groundwater is highly contaminated with arsenic and acts as the primary source of arsenic poisoning among the peoples of those areas. The results indicate that human population is affected with arsenic locally using the contaminated water for a long time.

Key words: *Arsenic, groundwater, Bangladesh, gamma ray activity, neutron activation analysis, trace elements*

INTRODUCTION

Arsenic is one of the most commonly studies trace elements as it is both toxic and carcinogenic (Nriago, 1994). It is reported that 40 % of arsenic in human body comes from food chain (BIAM, 2002). Arsenic has been taken into account as a dangerous environmental pollution and detected as a serious health risk in many countries of the world, reported from Argentina, Mexico, Chile, USA, Taiwan, Mongolia, Thailand, Philippines, China, Japan, India, as well as Bangladesh (Akram, 1997, BGS, 2001 and SOES-DCH, 2000). The arsenic contamination is not only a health hazard for the people; it also affects the environment and creates social problems (Table 1). There are two main theories (Pyrite oxidation and oxyhydroxide reduction) as to how arsenic is released into the ground water.

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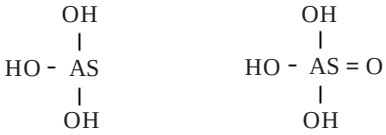
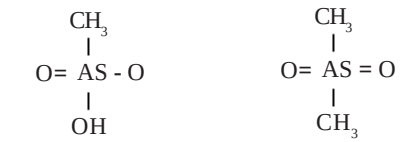
Toxic, naturally occurring arsenic species



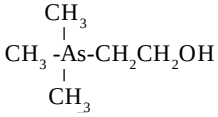
Arsenic III Arsenic V

Metabolic by products of arsenic V

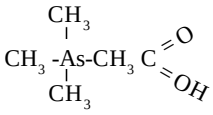


Monomethyl arsenic Dimethyl arsine

Non-toxic species of arsenic in food products



Arsenocholine



Arsenocbetaine

The mechanism is illustrated in Fig. 1.

MATERIALS AND METHODS

Study area

Feni district of Bangladesh is selected for this experiment. It has six upazila. Each upazila comprises of several Unions. Each Union has some 'blocks' (wards). In each 'blocks' there are some 'Villages'.

Table 1: Arsenic polluted countries with affected population and main reason

Name of the country	Year of pollution	Population affected by arsenic	Maximum range of pollution (mg/L)	Main reason of pollution
Argentina	1938–81	20,000	0.1–2.0	Natural soil pollution
Mexico	1963–83	200,000	0.1–0.5	Oxidation of arsenic bearing minerals
Chile	1957–69	130,000	0.8–1.3	River cutting through arsenic bearing formation
USA	1972–82	3,000,000	0.045–0.092	Oxidation of pyrite, reduction of ferric oxide, etc.
Taiwan	1961–85	100,000	>0.05	Oxidation of pyrite
Mongolia	1962–89	1,774	>0.05	Over-irrigation
Thailand	1987–98	18,000	0.05–5.0	A tin mine
Philippines	1992–95	39	-	Geothermal power plant
China	1953–93	1,546	-	Use of coal as fuel
Japan	1945–95	217	-	Metal and coal mine
India	1978–98	200,000	0.05–3.7	Over-exploitation of groundwater (pyrite oxidation)

(Akram, 1997; BGS, 2001 and SOES-DCH,2000)

Table 2: Statistics of Arsenic contamination with affected areas and population in Bangladesh

Parameter	BGS (1999)	SOES-DCH (2000)
Total area of Bangladesh	147,570 km^2	147,570 km^2
Area of affected districts	134,275 km^2	112,407 km^2
Total population of Bangladesh	29.6 mL	129.6 mL
Population of affected districts	128.4 mL	93.5 mL
Total districts in Bangladesh	64	64
Surveyed districts	61	64
Districts having arsneic above max. permissible limit	61	47
affected thana	465	357
affected thana surveyed	411	222
having arsenic >0.05 mg	270	147
exposed population	80.1 mL	46.2 mL
Population who drink water with arsenic >0.05 mg/L	21 mL	25 mL

Table 3: Comparison of arsenic concentration determined in the standard reference material As (NIST, USA), Coal Fly Ash, Pond Sediment (NIST, USA), and SL-1, Soil-7, (IAEA) with the certified value

Standard Sample	Element	Experimental value μg/g (ppm)	Certified value μg/g (ppm)	Deviation %
Soil-7	As	12.8 ± 0.7	13.4 ± 0.9	4.4
SL-1	As	29.7 ± 1.64	27.6 ± 2.9	6.8
As standard	As	19.6	20	2
Coal fly ash	As	140.5 ± 10	145 ± 15	3.2
Pond sediment	As	12.4 ± 1.29	12 ± 2	3.3

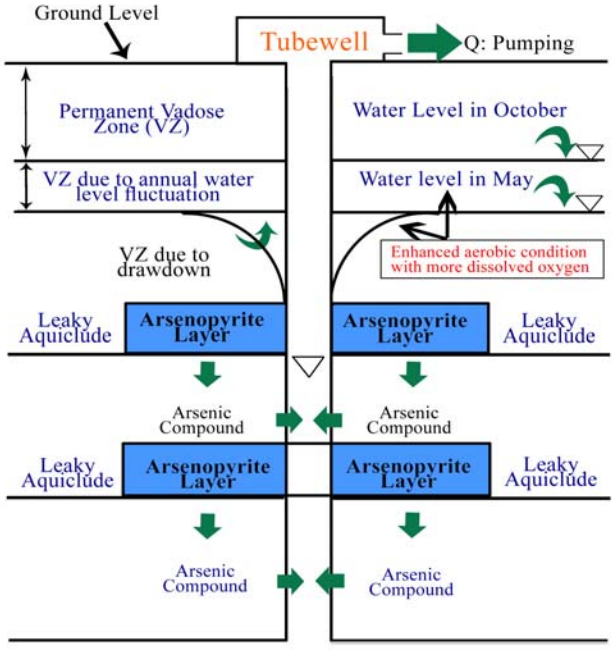


Fig. 1: Aerobic condition in groundwater around a tubewell

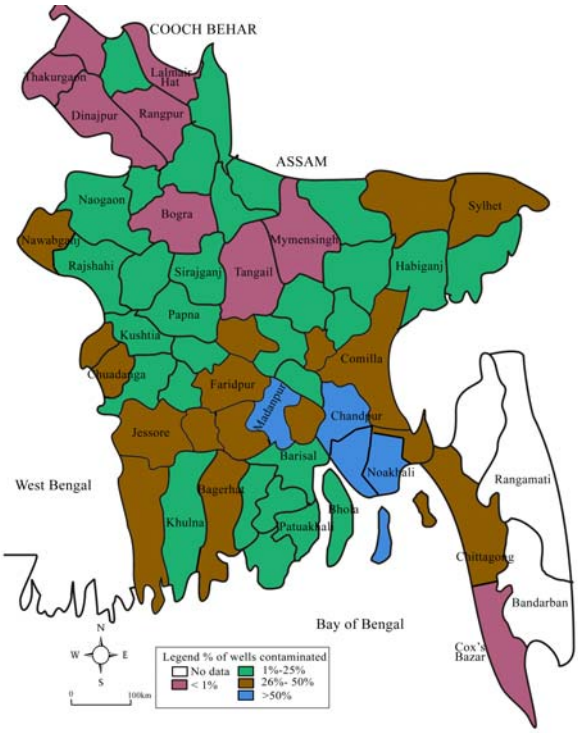


Fig. 2: Groundwater arsenic contamination in different districts of Bangladesh based on compilation of field and laboratory test data

Sample collection

Hair samples were collected from the tube well water users. Hair samples of the water users of this district have been categorized into two: below 12 years of ages and upper 12 years and are coded as Hb and Hu, respectively. Before, collecting samples, scissor was cleaned by acetone to avoid the others contamination.

Sample preparation

Hair samples were transferred from polyethylene bag to the Petri dish. In order to clean hair samples properly, samples were washed and dried in the following ways;

i) Acetone and dried naturally
ii) Deionized water and then dried
iii) Again by acetone and dried

The dried clean hairs were cut into small pieces. Four types of standard reference materials were used for the analysis of two sets of hair samples (a total of 30 samples) supplied by National Institute of Standard and Technology (NIST). Whereas the first set was analyzed using the International Atomic Energy Agency (IAEA) certified reference materials Soil-7 and SL-1, the second set was analyzed using coal fly ash and pond sediment. About 100 mg hair was taken into the bullet type irradiation vial. The identification of the sample was put at the outer surface of the vial and heat-sealed.

Irradiation and data counting

Al-Au Flux monitors were used between the two set of samples to determine neutron flux gradients. The samples and standards were irradiated in the 3 MW TRIGA MARK-II research reactors at the Atomic Energy Research Establishment (AERE), Savar, and Dhaka, Bangladesh. The irradiation was performed with neutron flux of 1.52×10^{13} n/cm^2/sec at a power level of 500 kW for 20 min. using Dry Central Thimble (DCT) facility, counting time for hair was 3600 sec. Below Fig. 3 show the irradiation capsule, get ready for irradiation.

After irradiation completes the samples, standards and flux monitors were counted with a high purity germanium detector coupled with a personal computer based S-100 Multi Channel Analyzer (MCA) master board packages. The energy spread in multichannel analyzer was adjusted to 0-1500 keV over 4096 channels.

Gamma ray counting were performed for samples and standards after a decay time of 1-2 day, with a measuring time of 500-3600s for the determination of As [$^{75}As(n,\gamma)^{76}As \rightarrow ^{76}Se$ ($t_{½}$ = 26.3 h; E_γ = 559.1 keV)]. Countings for flux monitors were performed with a measuring time of 60s for the determination of Au [$^{197}Au(n,\gamma)^{198}Au$($t_{½}$ = 2.35 d; E_γ = 411.6 keV)]. The dead time was kept below 7% during counting for both samples and standard.

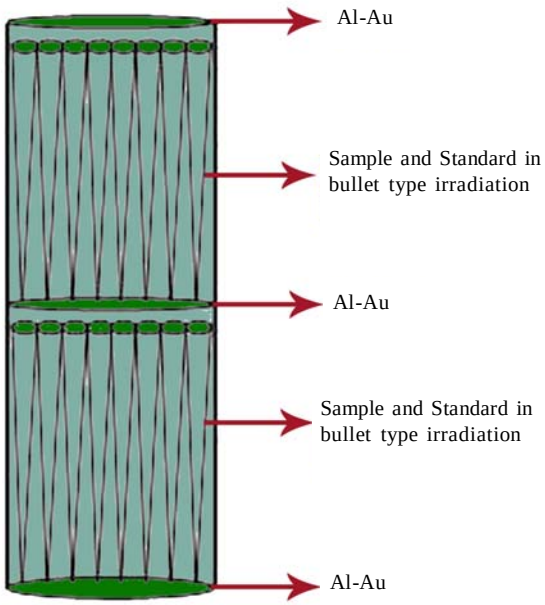


Fig. 3: An irradiation capsule gets ready for irradiation.

Calculation of arsenic concentration

For the calculation of concentrations, the peakgr10 software package was used. Concentratrions calculation were also performed manually by the following activation equation

$$\frac{\text{Weight of element "a" in sample}}{\text{Weight of element "b" in std.}}$$

$$= \frac{A_{x^*} \text{ in sample } e^{+\lambda t \text{ sample}}}{A_{x^*} \text{ in } .e^{+\lambda t \text{std.}}}$$

Where, 'a' and 'b' are the weight of the element in the sample and standard respectively. A_{x^*} is the activity in sample and standard (Laul 1979).Both the program and manual calculations gave the consistent results.

Accuracy of the analysis and the total uncertainty

Quality Assurance (QA) test is performed to investigate the reliability of the analysis by measuring arsenic concentration level in standard reference materials As (NIST, USA), coal fly ash, SL-1 and Soil-7 (IAEA) relative to primary standard of arsenic. The analytical results for certified reference materials are given in the table below. The experimental results are within 10% deviation, which is a good agreement with the certified value. This deviation was achieved due to poor counting statistics. Uncertainties of the samples were calculated according to the 1993 ISO guide and were added to the uncertainty in the measurement (Kuèera, *et al.*, 1999). Table 3 Shows that Comparison of arsenic concentration determined in the standard reference material As (NIST, USA), Coal Fly Ash, Pond Sediment (NIST, USA), and SL-1, Soil-7, (IAEA) with the certified value.

RESULTS

The obtained results of arsenic concentration in the lower age Hb (below 12 years of age users) categories of users is in the range of 0.33 to 3.29 μg/g and that in the Hu (upper 12 years of age users) categories is in the range of 0.47 to 6.64 μg/g . Arsenic detected in the samples Hb-1 to Hb-6 is in the range of 0.13 to 3.29 μg/g , where as arsenic in corresponding upper age users samples Hu-1 to Hu-6 is in the range of 1.48 to 6.64 μg/g . Arsenic in the samples Hb-7 to Hb-14 is in the range of 0.33 to 1.2 μg/g , where as in corresponding upper age users samples Hu-7 to Hu-14 is in the range of 0.24 to 5.75 μg/g. Maximum permissible range of arsenic in the human scalp hair is 1 μg/g certified by WHO.

DISCUSSION AND CONCLUSION

In this study, arsenic is detected to the human scalp hair samples for both the Hb and Hu categories. It is shown in Fig. 4 that arsenic is detected in human scalp hair for both the Hb (below 12 years users) and Hu (Upper 12 years users) categories, but from the above Figs it is shown that the under 12 years old peoples (Hb) are less affected compared to the more than 12 years old peoples (Hu). From Fig. 4, it is shown that the samples Hb-1 to Hb-6, which were collected from the areas of the Sonagazi, Dagon bhuiyan Upazilas of the district, show and lower accumulation of arsenic where as from Fig. 4, the samples Hu-1 to Hu-11 show

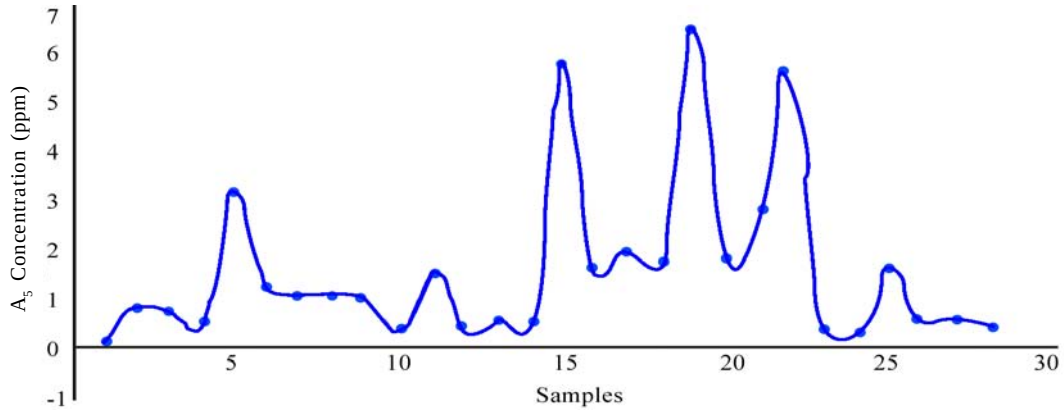


Fig. 4: Arsenic Concentration in human hair scalp hair

the higher accumulations. Hair samples Hb-7 to Hb-12 show very high accumulation of arsenic and the groundwater in the corresponding areas were found highly contaminated by arsenic and is in the range (110-190) mg/L (Uddin, *et al.*, 2005). Hair samples Hb-13 to Hb-14 in the under 12 years category show the lower accumulations of arsenic (0.45-0.63)μg/g which is justified to the corresponding lower arsenic found in the groundwater in the respective areas and is in the range (5-43) μg/g (Uddin, *et al.*, 2005). Results were obtained from the experiment that the human hair samples, the fewer than 12 years old users (Hb) are less affected compared to the more than 12 year old users (Hu).

Therefore, it is concluded that higher values of arsenic in hair samples were due to the longtime accumulation of arsenic through the uses of contaminated groundwater. Results show that the ground water is contaminated with arsenic and acts as the primary source of arsenic poisoning among the peoples of those areas. The results indicate that human population is affected with arsenic locally using the contaminated water for a long time.

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