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Detection of Arsenic in Water, Herbal and Soil Samples by Neutron Activation Analysis Technique

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Abstract Arsenic contamination of ground water is well understood while other environmental systems are rarely considered to be contaminated by arsenic. A vital issue is whether or not appreciable arsenic transmits through the food chain. Reportedly, ayurvedic herbal medicine products (AHMPs) manufactured in Asia were found to be contaminated by harmful level of Arsenic. This study was aimed to quantify the arsenic levels in water, herbal and soil samples collected from the same origin using highly accurate neutron activation analysis (NAA) technique. Harmful level of arsenic was detected in most of the water and soil samples. Moreover, a considerably harmful level of Arsenic was detected in herbal samples collected from the same origin. As a result, AHMPs manufactured in Asia might be contaminated by arsenic through arsenic contaminated herb plants.

Keywords Arsenic · Herbal · Neutron activation analysis (NAA) technique

Arsenic (As) contamination of ground water is a serious threat to public health. More than 100 million people worldwide have been estimated to be chronically exposed to As from drinking water containing high As levels (Alaerts et al. 2001; Chowdhury et al. 2000; Dhaka Community Hospital Trust 2005). Of the various sources of arsenic in the environment, drinking water probably poses the greatest threat to human life (British Geological Survey 1998). Though many countries have reported to be affected by As contaminated drinking water, the situation is perhaps most devastating in Bangladesh because of the number of affected people of its large population [Bangladesh Arsenic Mitigation Water Supply Project (BAMWSP) 2006; Josephson 2002]. Arsenic is also found in soil, vegetables and other crops such as snake gourd (As level, 0.489 mg/L), ghotkol (0.446 mg/L) (Alam et al. 2003). Harmful levels of arsenic have also been found in the ayurvedic herbal medicine products (HMPs) manufactured in Asia. One of five Ayurvedic HMPs produced in South Asia and available in Boston South Asian groceries stores contains potentially harmful levels of Arsenic (Saper et al. 2004). The exact time of onset of As exposure in Bangladesh is unknown, it is suspected to have started during the 1960s and 1970s when the United Nations Children's Fund (UNICEF), in collaboration with the Bangladesh government, started to install hand-pumped tube wells to provide pathogen-free drinking water to the population (Smith et al. 2000). However, the contamination of As in tube-well water in Bangladesh was clearly identified in 1993 by the Department of Public Health Engineering in the Chapai Nawabganj district in the northwestern part of the country. According to the latest surveys at least 53 of the 64 districts of Bangladesh are affected with Arsenic pollution (Dhaka Community Hospital Trust 2005). Until now there are not many systematic research on the measuring the exact levels of arsenic in water, soil or plant samples.

This study aims to assess the distribution and levels of arsenic contamination in three environmental components:

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water, soil and herb plants by using neutron activation analysis technique.

Materials and Methods

Tube-well water, soil and plant samples were collected from different locations in Narayanganj District, Bangladesh. A house-to-house survey was conducted and the tube-well owners were interviewed. The soil and plant samples were collected from the surrounding area of the tube-wells from which the water samples were collected. Only the surface soil samples were collected.

The collected water samples were preserved with 37% hydrochloric acid. Five milliliter of 37% hydrochloric acid was added to each 100 ml water sample. With a fine pipette, 500 µl of the solution was dropped on an eightfold filter paper. The water soaked filter paper was then weighed in a microbalance to determine the weight of the water. The filter paper was then allowed to dry after the water was absorbed. It was then wrapped in a twofold polyethylene paper and heat-sealed. Following the same procedure, 20 µl of 1,000 mg/L IAEA arsenic standard reference material and 200 µl of 4.42 mg/L IAEA arsenic standard material were prepared for irradiation.

Soil samples were collected from the rooting zone of the plants surrounding the tube wells where the water samples were collected. The samples were transported in dried acetone-cleaned polyethylene bags (film container). The samples were then transferred from the polyethylene bags to the acetone-cleaned Petri dishes. They were then dried in an oven at 70°C until a constant weight was achieved. Each dried sample was grounded to homogeneous powder in an agate motor separately. The grounded samples were stored in separate polyethylene packets as stock samples. Each polyethylene packet had 55 mg. Similarly, the IAEA certified reference materials -soil-7 and lichen were placed in clean small polyethylene bags (similar weight); the bags were coded on the outer surface. The bags were heat sealed and then encapsulated in polyethylene papers, which were also heat-sealed. The samples were divided into several sets. Each set consists of a collected sample and two types of the IAEA reference materials. Each set was placed in an irradiation vial in preparation for irradiation.

Leaves were collected from plants (basil) surrounding the tube-wells from which the water samples were collected. The plant samples were prepared for irradiation following the same procedure described above.

The samples were analyzed by the reactor based (TRIGA MARK-II Research reactor) Instrumental Neutron Activation Analysis (INAA) method. Standardization was achieved by relative method. The tested samples and the standard reference materials were irradiated in the dry central thimble (DCT) facility of three MW TRIGA MARK-II research reactor at the Institute of Nuclear Science and Technology (INST), Atomic Energy Research Establishment (AERE), Dhaka, Bangladesh with thermal neutron flux 7.6×10^{12} n/cm^2/s.

Table 1 Comparison of experimental values with the certified values (in mg/L)

	As std. lichen	As std. water	As std. water
Present work	0.51	4.66	1,011
Certified value	0.517	4.42	1,000
RSD%	1.4	5	1.1

The NAA is used in many biological and environmental researches. It is highly accurate and is also used to validate the results of other analytical methods (Alfassi 1994). It was used in detecting arsenic in toenails and well water samples by Schmitt et al. (2005), deparaffined skin tumor samples of arsenic-related disease by Matsui et al. (1999) and brain tumor tissue (Zhuang et al. 1996).

The Instrumental NAA was standardized with a relative method. The collected samples and standard reference material were irradiated in the Dry Central Thimble (DCT) facility of three MW TRIGA MARK-2 research reactors with thermal neutron flux of 7.6×10^{12} n/cm^2/s. The γ-rays emitted from the irradiated samples were detected with a HPGe detector and analyzed with a computer based Canberra S-100 multi channel analyzer (MCA). The accuracy and precision of the results were checked by applying the same experimental protocols as that of the standard reference materials (SRMs): IAEA-As std. Soil-7, IAEA- As std. (Lichen), As std. 1,000 mg/L and As std. 4.42 mg/L water. The relative standard deviations (RSD %) of the certified values of the standard reference materials (SRMs) from the experimental values are shown in Table 1.

Lead was used to shield the HPGe detector from cosmic radiation and the natural radioactivity in the environment. The resolution of the HPGe detector was determined with a calibrated ^{60}Co point source, which was placed on the surface of the detector. The resolution was found to be 1.94 keV at the 1,332.51 keV gamma line of ^{60}Co source (IAEA 1998). The detection limits (DLs) of arsenic concentrations were determined using the lowest concentrations found in water, soil and herbal samples. The findings of arsenic concentrations were well above the DLs.

Results and Discussion

The highest levels of arsenic concentrations 0.20, 11.35 and 0.79 mg/L were found in water, soil and plant samples

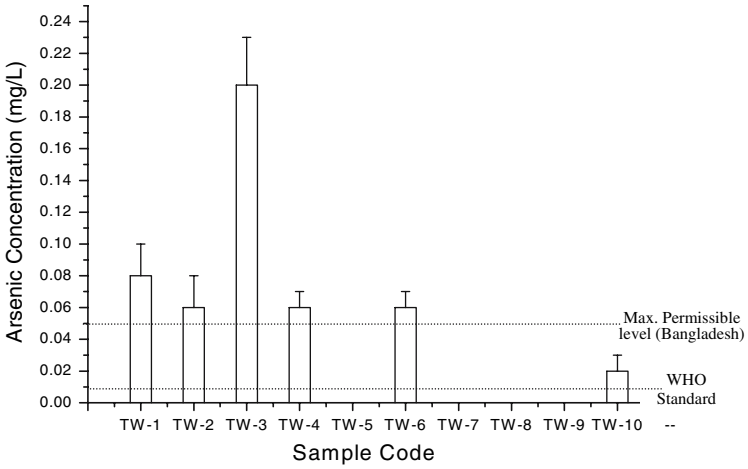

Fig. 1 Arsenic concentration in water samples collected from Narayanganj, Bangladesh

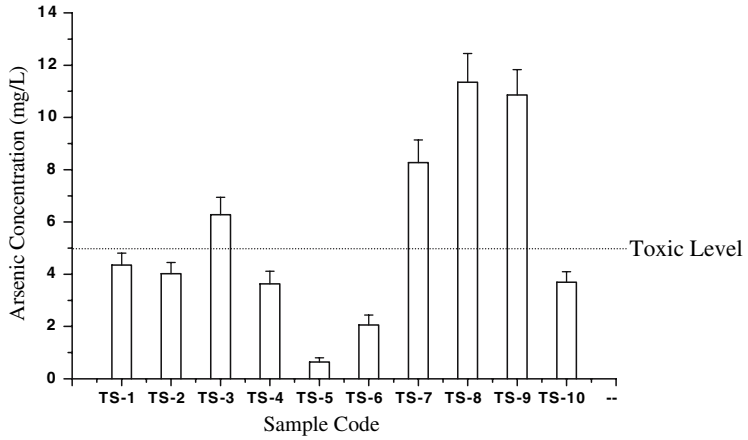

Fig. 2 Arsenic concentration in soil samples collected from Narayanganj, Bangladesh

respectively. The arsenic concentrations in individual collected samples and the maximum permissible level are shown in Fig. 1. The maximum level (0.20 mg/L) of arsenic was observed in water sample TW-3 (Fig. 1). Arsenic was detected in six of ten water samples, only one of which was within the WHO permissible limit. All but one-soil samples (TS-5) were found to be contaminated by arsenic (Fig. 2). Five of ten herbal samples were contaminated by arsenic (Fig. 3)

High arsenic contamination was demonstrated in the tube-well water of the Sonargaon region. Six out of the ten water samples were contaminated ranging from 0.02 to 0.20 mg/L. The US Environmental Protection Agency (US EPA) standard for As in drinking water is 0.05 mg/L (US EPA 1993). The WHO maximum permissible limit for arsenic in drinking water is 0.05 mg/L. The WHO recommended guideline value of arsenic in drinking water is 0.01 mg/L (World Health Organization 1992). In 1996, the WHO reported that in 11 districts in Bangladesh, arsenic levels in ground water were at high concentrations in drinking water and that 23 million people were at risk. Chowdhury et al. (2000) reported that in 42 districts arsenic levels in ground water were at high concentrations. A national survey (Bangladesh Arsenic Mitigation Water Supply Project 2006) reported that amongst the country's 7–11 million hand-pumped tube wells, approximately half have been estimated to supply groundwater with an As concentration >0.05 mg/L. Our study reports around 50% of tube well in Narayanganj exceed the WHO maximum permissible level, and only 10% were below the permissible level. Therefore the tube well water in this region is not suitable for drinking, cooking and agricultural purposes.

The arsenic concentrations in soil ranged from 0.63 to 11.35 mg/L. The normal range for As in soils of various countries was 0.1–40 mg/L (0.1–40 mg/kg) (Chatterjee et al. 1993). It was indicated by a conservative risk analysis that As concentration in soil could reach 4 mg/L without becoming a hazard to exposed organisms. The As concentration in the soil quality criterion set by US EPA (Environmental Protection Authority) for total arsenic is 2 and 5 mg/L in soil is toxic. Our finding reports that all but one soil sample exceeded the maximum permissible level of arsenic in soil. Therefore crops grown in soil in this region are serious health hazards.

The arsenic contents in plant (basil) samples ranged from 0.18 to 0.79 mg/L. Basil leaves are used in Ayurvedic

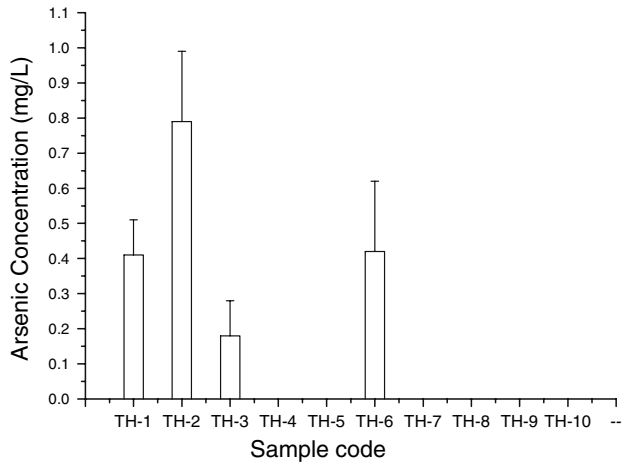

Fig. 3 Arsenic concentration in herbal samples collected from Narayanganj, Bangladesh

medicine. The traditional healthcare systems practiced in Bangladesh include the Ayurvedic, Unani, Homeopathic, and Folk medicine systems. Ayurvedic system is one of the oldest systems of medicine, which has been practiced in this subcontinent for over 3,000 years (WHO 1976).

As basil leaves are used in Ayurvedic Medicine, manufacturers should take necessary precautions to avoid using arsenic contaminated plants. Therefore the harmful presence of arsenic in Ayurvedic Herbal Medicine Products (AHMPs) may be due to absorption of the arsenic contaminated water. However, in our study, the arsenic contents in herbal and water samples were not proportionate.

The strength of this study is the use of neutron activation analysis technique to quantify the As level more accurately. The major limitation of this study is the small sample size. However, measuring the arsenic levels in the standard reference materials quality assurance (QA) was performed. The results are within 5% deviation from that of the certified values, which is considered acceptable in statistical point of view where 5% error is tolerable.

In conclusion, approximately 50% tube wells of Narayanganj District, Bangladesh exceeded the permissible level and 40% of soil samples around the tube wells had more than 5 mg/L arsenic which is toxic. Basil plants had arsenic if they were grown in arsenic contaminated water. Our study suggests that an assessment of arsenic concentration by using neutron activation analysis technique with a large number samples will provide more accurate measure in water, soil or other environmental systems.

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