



Transformation of arsenic in the presence of cow dung and arsenic sludge disposal and management strategy in Bangladesh



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SUMMARY

With increasing use of arsenic (As) removal units for treatment of As-contaminated groundwater in rural Bangladesh, concerns have been raised regarding safe disposal of the As-rich wastes from such units and possible contamination of the environment. In the absence of any clear guideline for safe disposal of wastes generated from As removal units, the wastes are usually disposed of in the open environment, often on cow dung beds in the backyard. Short term (up to 6 weeks) batch experiments performed in this study suggest that bio-chemical (e.g., bio-methylation) processes in the presence of only fresh cow dung may lead to a significant removal of As, both from aqueous solution and As-rich treatment wastes. Arsenic removal appears to increase with decreasing As to cow dung weight ratio. This study also suggests that arsenate transforms to arsenite before removal from aqueous As solution in the presence of cow dung. In most cases majority of As removal takes place during first few days. Removal of As under cap-open (to facilitate aerobic condition) and cap-closed conditions (to facilitate anaerobic condition) were found to be similar. No significant variation was observed in the removal As from aqueous solution and from treatment wastes (As bound to iron solids). This study concludes that disposal of As-rich treatment wastes to cow dung pits could be an effective option of As sludge disposal and management in rural areas of Bangladesh.

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1. Introduction

Widespread arsenic (As) contamination of groundwater has become a major threat to potable water supply sector in Bangladesh. Groundwater extracted from shallow aquifer with hand-tubewells is the primary source of drinking water for majority of its population of about 160 million, especially those living in the rural areas. An estimated 270 Upazilas (sub-districts) out of 465 have been affected with significantly high concentrations of As (BGS and DPHE, 2001; Ahmed et al., 2004; BAMWSP, 2005). An estimated 35 million people are exposed to arsenic concentrations in tubewell water above the Bangladesh drinking water standard of $50 \mu\text{g L}^{-1}$ and 57 million people are above the WHO guideline value of $10 \mu\text{g L}^{-1}$ (BGS and DPHE, 2001; Gaus et al., 2003; Maddison et al., 2005). It has been demonstrated that As is a potent carcinogen (Morton et al., 1976; Duker et al., 2004); close to 40,000 cases of arsenicosis have been detected in a nationwide survey in Bangladesh (BAMWSP, 2005; UNICEF, 2010), and public health experts believe there will be more than 2.5 million cases in the next 50 years (Russell, 2006).

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For providing safe water in the As affected areas, a number of household and community based As removal systems have been and are being developed and a number of arsenic removal systems are currently being used in many arsenic affected areas. All As removal systems generate some form of As-rich waste. The volume of waste, and concentration and chemical stability of As in the waste depend on the concentration of As in the raw water and the treatment technology used. Broadly, the As-rich waste materials can be classified into: (a) wastes generated from coagulation/precipitation based systems, and (b) wastes generated from systems based on adsorptive filtration and other techniques (e.g., ion exchange) (Ali et al., 2003; Sullivan et al., 2010). The waste belonging to the first category is primarily slurry containing coagulated flocs of alum or iron salt, rich in arsenic. The waste belonging to the second category is primarily spent adsorbents or ion exchange resins, rich in arsenic (Ahmed, 2001; Rahman, 2004). Sullivan et al. (2010) reported lack of effective As sludge management practices in Bangladesh and India. In the absence of any clear guideline for safe disposal of wastes generated from As removal units, the wastes are usually disposed of in the open environment, often in a cow dung bed in the backyard of typical homes in rural Bangladesh. With increasing use of As removal units, concerns have been raised regarding safe disposal of these wastes and possible contamination of environment from arsenic present in the

wastes. However, there is only limited data on the quantities and characteristics of these wastes (Hamel and Zinia, 2001; Sullivan et al., 2010) and possible mobilization of As from these wastes (Badruzzaman, 2003; Ali et al., 2003).

Biochemical transformation of As plays an important role in determining fate of arsenic in arsenic-rich treatment wastes. Microbes can transform arsenic present in biosolids (sludge) (Mukhopadhyay et al., 2002; Islam et al., 2005; Li et al., 2011; Yin et al., 2011) into soluble and volatile species through the processes of oxidation, reduction and methylation (Anderson and Bruland, 1991; Huang et al., 2012). Inorganic As can be metabolised to both volatile organic species, such as monomethylarsine (MMA), dimethylarsine (DMA), and trimethylarsine (TMA) and nonvolatile species such as methylarsonic acid, dimethylarsinic acid, and trimethylarsenic oxide (Bentley and Chasteen, 2002; Lloyd and Oremland, 2006; Ye et al., 2012). Biomethylation of inorganic As by yeast, fungi and bacteria cause the release of gasified arsenic into the atmosphere from the earth surface and volatile As is usually less toxic (Tamaki and Frankenberger, 1992; Edvartoro et al., 2004). In this detoxification mechanism As(V) reduces to As(III) (e.g., Dowdle et al., 1996; Nikolic, 2008) and microbial activity can speed up the reduction process (Ahmann et al., 1994; Lloyd and Oremland, 2006).

Cow dung is a widely available marvellous resource that is constantly being produced and deposited in large quantities in rural areas in Bangladesh. Even though it has passed through an animal's digestive tract, dung retains many nutrients (Smith and Wheeler, 1979). Thus, it attracts bacteria, fungi, protozoa, platyhelminths, nematodes, annelids, and arthropods (Hauser, 1994), which further enhances natural biogeochemical processes (Adegunloye et al., 2007). Researcher detected bacterial genera e.g. *Escherichia coli*, *Salmonella coliform* bacteria (Gong, 2007; Geetha and Fulekar, 2008), *Acinetobacter* sp., *Aliccaligenes* sp., *Bacillus* sp., *Pseudomonas* spp. *Setratia* spp. *Nocardia* sp., etc. in cow dung (Geetha and Fulekar, 2008; Akinde and Obire, 2008). Among them *Aliccaligenes* sp., *Nocardia* sp., *Pseudomonas* spp., *Escherichia coli* are capable to gasify arsenic (Shariatpanahi et al., 1981; Tamaki and Frankenberger, 1989; Rosen, 2002). Islam et al. (2005) found that the numbers of both As methylating bacteria and methanogens, which are most likely to be involved in arsenic methylation, are higher in cow dung than in soil. Mohapatra et al. (2008) reported volatilization of As by methanogenic bacteria from a system where cow dung was used as the major substrate.

Though little is known about the transformation of As into volatile forms in the presence of only cow dung, many researchers have suggested disposing the wastes from arsenic removal system on cow dung bed (e.g., Hwang, 2002; Islam et al., 2007; Sarkar et al., 2011). This paper assesses the transformation of As present in aqueous solution and in As-rich wastes generated from coagulation based As removal systems in the presence of only fresh cow dung.

2. Materials and methods

2.1. Collection of arsenic-rich treatment waste and cow dung

In this study, arsenic-rich wastes have been collected from two different types of coagulation-based As removal systems: (i) STAR bucket treatment unit (based on iron salt coagulation), and (ii) UNU (United Nations University)–BUET (Bangladesh University of Engineering and Technology) bucket treatment unit (based on ferric chloride coagulation). In both cases, the wastes consisted primarily of slurry containing As-rich iron precipitates. All samples have been collected from operational arsenic removal units. Wastes from STAR bucket treatment units were collected from

Laksmipur district, while those from UNU–BUET unit were collected from Adda village in Barura Thana of Comilla district. Both Laksmipur and Comilla districts are located in south-central Bangladesh and have been heavily affected by the arsenic-contamination of groundwater.

Fresh cow dung samples were collected in a polythene bag from an animal farm at Jatrabari Area of Dhaka, Bangladesh.

2.2. Experimental methods

2.2.1. Transformation of arsenic present in aqueous solution and in arsenic-rich waste

In order to assess transformation of aqueous As [both As(III) and As(V)] in the presence of cow dung, batch tests were conducted in 15-mL falcon centrifuge tubes. Aqueous solution of As(III) was prepared with standard As(III) solution (1000 ± 10 mg/L as As^{3+} , prepared from As_2O_3 ; HACH Co., USA); As(V) stock solution was prepared by dissolving required quantity of $Na_2HAsO_4 \cdot 7H_2O$ in deionized water. Aqueous arsenic solutions (12 mL) with known concentrations of either As(III) or As(V) were taken in a series of centrifuge tubes (a total of 8–10 tubes for each set of experiment). Fixed quantities (1, 2, 4, 6 or 8 g) of fresh cow dung were added to each of the tubes. The experiments were carried out under both cap-open (to facilitate aerobic condition) and cap-closed conditions (to facilitate anaerobic condition). One set of tubes was capped after purging N_2 gas and a similar other set was left uncapped. Nutrient or seed for micro-organisms was not supplied to the tubes. After particular intervals, one tube from each set was taken and centrifuged to separate the solid (cow dung and waste) from the liquid. The pH of the liquid was then measured with a pH meter. The liquid portion from each tube was then separated by decanting and its arsenic concentration was measured (after acidification). Arsenic content of the solid (cow dung and waste) portion was determined after digestion with aqua regia. Thus total arsenic present in the tubes was determined as a function of time (2–36 days) to monitor any change in total arsenic content in the presence of cow dung.

In order to assess transformation of arsenic present in treatment wastes in the presence of cow dung, experiments similar to those described above were conducted with the slurry wastes collected from both STAR and BUET–UNU bucket treatment units. First, arsenic contents of well-mixed slurry wastes were determined after digestion with aqua-regia. For batch experiments, fixed volumes of slurry wastes (5 mL for STAR wastes and 30 mL for BUET–UNU waste) were taken in a series of 50 mL falcon centrifuge tubes and fixed quantities of freshly collected cow dung were added to each tube. Then the same procedure as described above was followed.

2.2.2. Transformation of As(V) to As(III) in the presence of cow dung

For assessing transformation of As(V) present in aqueous solution into As(III) in the presence of cow dung, batch tests were conducted in 15 mL falcon centrifuge tubes. Aqueous arsenic solutions (12 mL) with known quantities (varying from $1.05 \mu\text{g}$ to $4.25 \mu\text{g}$) of As(V) were taken in a series of tubes. One gram of fresh cow-dung was added to each of the tubes. The tubes were left uncapped. After particular intervals, one tube was taken out at a time and centrifuged to separate the solid (cow dung) from the liquid. The liquid portion from each tube was then separated by decanting and a 2 mL portion was taken with a micropipette to measure total arsenic concentration (after acidification). The remaining liquid portion was then taken in a centrifuge tube and analysed for As(V) and As(III). For this purpose, the liquid in the centrifuge tube was acidified with $24 \mu\text{L}$ of ultra-pure HCl to lower the pH to 2.7–3.0. One gram of Biorad resin was then added to the solution and the tubes were shaken for 1 min. It was then allowed to settle for 4–5 min,

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