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Arsenic in Bangladeshi soils related to physiographic region, paddy management, and mirco- and macro-elemental status

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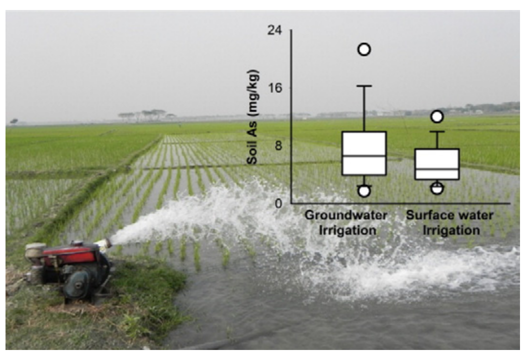
HIGHLIGHTS

- Arsenic accumulation in rice poses a human health risk.
- A large scale analysis across Bangladesh to determine soil arsenic concentration
- Soils from Holocene floodplains have higher soil arsenic concentrations.
- Paddy soils have elevated arsenic concentration compared to non-paddy soils.
- Surface water irrigated soils have lower arsenic compared groundwater irrigated soils.

GRAPHICAL ABSTRACT



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ABSTRACT

While the impact of arsenic in irrigated agriculture has become a major environmental concern in Bangladesh, to date there is still a limited understanding of arsenic in Bangladeshi paddy soils at a landscape level. A soil survey was conducted across ten different physiographic regions of Bangladesh, which encompassed six types of geomorphology (Bil, Brahmaputra floodplain, Ganges floodplain, Meghna floodplain, Karatoya-Bangali floodplain and Pleistocene terrace). A total of 1209 paddy soils and 235 matched non-paddy soils were collected. The source of irrigation water (groundwater and surface water) was also recorded. The concentrations of arsenic and sixteen other elements were determined in the soil samples. The concentration of arsenic was higher in paddy soils compared to non-paddy soils, with soils irrigated with groundwater being higher in arsenic than those irrigated with surface water. There was a clear difference between the Holocene floodplains and the Pleistocene terraces, with Holocene floodplain soils being higher in arsenic and other elements. The results suggest that arsenic is most likely associated with less well weathered/leached soils, suggesting it is either due to the geological newness of Holocene sediments or differences between the sources of sediments, which gives rise to the arsenic problems in Bangladeshi soils.

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1. Introduction

Rice is elevated in inorganic arsenic compared to all other dietary staples (Meharg et al., 2009). Flooding of soils, as in paddy cultivation, leads to the mobilization of natural and anthropogenic inorganic arsenic stored in iron oxyhydroxide phases, caused by both the reduction of arsenic and iron under negative soil redox potentials (Meharg and Zhao, 2012). Paddy soils are managed through tilling, fertilization, and surface water and groundwater irrigation, with the latter often elevated in inorganic arsenic throughout large areas of Bangladesh (Huq et al., 2003; Meharg and Rahman, 2003; Roberts et al., 2007; Lu et al., 2009). Furthermore, arsenic can undergo a number of processes within paddy soils that lead to its subsequent loss such as partitioning to monsoonal floodwaters (Dittmar et al., 2007; Saha and Ali, 2007; Dittmar et al., 2010; Roberts et al., 2010), leaching to sub-surfaces (McLaren et al., 2006; Khan et al., 2009; Heikens et al., 2007), and biovolatilization to arsines (Mestrot et al., 2011). Thus, the arsenic loading of any particular paddy soil will be due to geological origin and the subsequent weathering of constituent minerals, and the agronomic management of that sediment (Lu et al., 2009).

Bangladesh has three major geomorphological units (Brammer, 1996; Huq and Shoaib, 2013). These are hill, terrace, and floodplain areas. The hills occupy 12% of the country's land area. The uplifted terrace areas are of Pleistocene age and occupy 8% of the country. The floodplains are of Holocene age and occupy 80% of the country. The Holocene floodplains include the piedmont plains, river floodplains, tidal floodplains, and estuarine floodplains. These geomorphological units are related to the parent geological formations, however, they are also characterised by land topography and age of the soil formation through sediment deposition over time (Brammer, 1996).

To understand and characterise the physiography of the geomorphological areas, Bangladesh is divided into twenty main physiographic regions (FAO/UNDP, 1988). This physiographic classification was based on the parent material in which individual soil types were formed and the landscape on which the soils were developed (FAO/UNDP, 1988). Therefore, the physiographic regions have differences in geology, relief, drainage, age of land formation and pattern of sedimentary deposition. These differences ultimately influence the nature and properties of the soils in the different physiographic regions.

The biogeochemical cycling of arsenic in soils is strongly affected by other elements. Iron is central due to the strong association between insoluble arsenate and iron(III) oxyhydroxides under aerobic conditions and with the mobilization of iron (II) and arsenite under reducing (that is, paddy) conditions (BGS/DPHE, 2001; Smedley and Kinniburgh, 2002; McArthur et al., 2004; Polizzotto et al., 2005). Manganese oxides also have a similar redox chemistry to iron and are strongly implicated in arsenic immobilization/mobilization during oxic/anoxic cycling of paddy sediments (Smedley and Kinniburgh, 2002; Hasan et al., 2007). Arsenate is a phosphate analogue and, thus, key to competition for binding sites within the soil solid phase, as well as having similar biogeochemical cycling under oxic conditions (Adriano, 2001; Meharg and Hartley-Whitaker, 2002; Smith et al., 2002; Lambkin and Alloway, 2003; Stachowicz et al., 2008). Calcium and magnesium immobilize arsenate under oxic conditions, and could also have a role in the biogeochemical cycling of arsenic at a landscape level (Smith et al., 2002; Stachowicz et al., 2008; Fakhreddine et al., 2015).

Here, we wanted to understand the relationship between soil arsenic and paddy management practice with respect to arsenic loadings in Bangladeshi soils. Cultivation zones of paddy soils ($n = 1209$) across ten physiographic regions of Bangladesh, from latitude 22°06′ to 24°53′, and longitude 88°20′ to 90°59′ were sampled and analysed for arsenic and a suite of sixteen other elements (aluminium, calcium, cadmium, cobalt, chromium, copper, iron, lead, magnesium, manganese, molybdenum, nickel, phosphorus, potassium, sodium, and zinc). For a subset of soils ($n = 235$), paired paddy and adjacent non-paddy soils were also collected and characterised. The data were used to address four specific objectives: to assess the impact that geomorphological differences have on soil arsenic at a landscape level; to understand the relationship between the concentration of arsenic in paddy soils with arsenic in the underlying groundwater; to determine if the source of irrigation water impacts on soil arsenic concentrations; and by examining the concentrations of arsenic and other elements in paddy and non-paddy soils, we aimed to understand the impacts that paddy management has on soil elemental concentrations.

2. Materials and methods

2.1. Collection of soil samples

A total of 1444 soil samples (topsoil, 0–15 cm from the surface) from paddy fields ($n = 1209$) and neighbouring non-paddy areas ($n = 235$) were collected from 10 different physiographic regions within 57 sub-districts (upazilas) from 17 districts of Bangladesh (Table S1). Non-paddy soils were defined as the soils where paddy cultivation and groundwater irrigation had not been practiced within known memory of the farmers. The physiographic regions from where the soil samples were collected included Arial Bil ($n = 42$ paddy and 10 non-paddy soils), Brahmaputra Floodplain ($n = 207$ paddy and 64 non-paddy soils), Ganges River Floodplain ($n = 261$ paddy and 58 non-paddy soils), Ganges Tidal Floodplain ($n = 47$ paddy and 11 non-paddy soils), Gopalganj-Khulna Bils ($n = 63$ paddy and 8 non-paddy soils), Karatoya-Bangali floodplain ($n = 15$ paddy soils only), Meghna Estuarine Floodplain ($n = 204$ paddy and 28 non-paddy soils), and Meghna River Floodplain ($n = 184$ paddy and 26 non-paddy soils) from Holocene floodplains, and Barind Tract ($n = 68$ paddy and 15 non-paddy soils) and Madhupur Tract ($n = 118$ paddy and 15 non-paddy soils) from Pleistocene terraces. The source of irrigation water for the paddy soils was recorded (groundwater, $n = 904$; surface water, $n = 281$; both, $n = 24$). Only the soils that had a non-mixed irrigation source were used for analyzing the impact of irrigation type on soil arsenic.

2.2. Sample processing and preparation for analysis

The soil samples were air-dried and, prior to analysis, the samples were oven dried (80 °C ± 5 °C for 48 h), and finely ground using a ball-mill. The soil digestion procedure followed was described by Adomako et al. (2009). Briefly, 0.1 g of soil was placed in a glass digest tube and 2.5 ml of concentrated nitric acid was added to the tube and left overnight for pre-digestion. Then, 2.5 ml of hydrogen peroxide was added to the sample just before digesting and the sample was heated on the block digester for 1 h at 80 °C, for 1 h at 100 °C, for 1 h at 120 °C, and finally, at 140 °C for 3 h until the solution was clear. Once cooled, the digested soil samples were transferred into 15 ml polypropylene tubes and each glass tube was thoroughly rinsed 3 times with ultrapure deionized water (Milli-Q 18.2 MΩ). The volumes were made up to 15 ml mark using the same water. To obtain the appropriate dilution for analysis by inductively coupled plasma-mass spectrometer (ICP-MS) and microwave plasma-atomic emission spectrometer (MP-AES), the samples were further diluted to 1 in 10. Calibration standards were prepared from 1000 mg/l multi-element stock solutions (SPEX CertiPrep Reference Material).

2.3. Chemical analysis

The pH of the soil samples were measured at a soil:water (deionized water) ratio of 1:2.5 (Huq and Alam, 2005). The ICP-MS (Agilent Technologies 7500c, Japan) was used to determine the total concentrations of arsenic, cadmium, cobalt, copper, chromium, lead, manganese, molybdenum, nickel, phosphorus, and zinc in the soil digests and the MP-AES (Agilent Technologies 4100 Series, USA) was used to determine the total concentrations of aluminium, calcium, iron, magnesium, potassium, and sodium in the soil digests. In each batch of digestion, 10% of