Impact of use of As-contaminated groundwater on soil As content and paddy rice production in Bangladesh

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Abstract: Arsenic (As) contamination of irrigation water and soils of Bangladesh was found to be highly variable at scales from the command area of a tubewell to nationwide. Spatial pattern in soil As at the command area scale was created as irrigation water was rapidly oxygenated and As adsorbed on precipitated ferric hydroxides. Complex relationships between irrigation water and soil As levels were found in an study of 5 upazilla. At the national scale, soil As was elevated in the Gangetic floodplain indicating deposition of As contaminated sediments from this river. The pattern of soil As concentrations was very different from that for irrigation water, which matched the published pattern for household tubewells. Arsenic was shown to be phytotoxic to all tested rice varieties in a farmer command area where there was a soil As gradient from 11-67 mg kg^{-1}. Production of rice in a more aerobic environment on raised beds was able to substantially prevent phytotoxicity. Rice from the national and Upazilla surveys was found to be elevated in As compared to a global "normal" range for As in rice. Raised bed production reduced As concentrations in rice straw and grain to 15-30% and 0-50%, respectively, of the values found with conventional paddy production. With increasing grain arsenic levels, rice from Bangladesh contained primarily inorganic As species, whereas rice from the USA increasingly contained dimethyl arsinic acid which is considered to be much less toxic to humans than inorganic As. At Bangladesh rice consumption rates, almost all Bangladesh rice would provide more inorganic As to adults than that allowed by the WHO drinking water standard of 10 µg L^{-1} and comparable amounts to that allowed by the Bangladesh drinking water standard of 50 µg L^{-1}.
Keywords: soil arsenic, irrigation water arsenic, Bangladesh arsenic contamination, environmental arsenic management, rice safety

1. Introduction

Contamination of shallow groundwater with arsenic (As) in Bangladesh was recognized in the 1990's [1]and has become a public health issue with the development of about 11 million household shallow tube-wells (STW's) nationwide. Many of these STW's in the central and southern regions of the country are accessing aquifers (30-70 m) containing more than 50 µg L^{-1} inorganic As. Beginning in the 1970's, Bangladesh government policy also facilitated a rapid development of irrigation tubewells and production of rice in the dry winter, or *boro*, season. The irrigation tubewells tapped the same shallow, potentially As contaminated, aquifers as the household tubewells. The development of *boro* season rice production was primarily responsible for the country attaining cereal self sufficiency and it now accounts for 55% of total rice production in Bangladesh [2]. Each crop of *boro* rice uses 1 to 1.5m depth of irrigation water that has the potential to add significant amounts of As to soil.

Inorganic arsenic species are retained in soils by adsorption on mineral oxide surfaces, with Fe-oxides generally considered to be the major sink for As in paddy rice soils when they are oxidized. Under the reducing conditions of the paddy, Fe-oxides dissolve and inorganic As is released into the soil-water matrix from which it can be assimilated by the growing rice plant. Uptake of As by rice is complicated by various chemical and physiological processes that occur in the rice paddy, namely (i) a change in oxidation state of As from arsenate (As-V) to arsenite (As-III) as reduction in the paddy intensifies [3-5], (ii) the formation of oxidized Fe plaque on rice root surfaces that readsorbs As, but in competition with phosphate [6-8], (iii) the possible formation of insoluble As sulfur species [9], (iv) competitive uptake of phosphate and arsenate through the same ion channel [10] (v) competitive uptake of arsenite and silicate through a general aquaporin channel [11,12] and (vi) microbial methylation of inorganic As to mono- and dimethyl-As species that have different rates of uptake than those of inorganic As species [10,13] Consequently we should expect that the levels and forms of As in soil solution and their uptake by rice will not be simply related to total soil As, or even available soil As, content which complicates establishment of a safe level of As in soils used for flooded rice production.

The uptake of As by rice and its translocation to rice grain has been assessed in a number of studies. In general, relative As concentrations in roots, foliage and grain decrease in the approximate ratio of 100:10:1. It should be recognized that As concentration in rice roots usually also includes that adsorbed on the Fe-plaque on root surfaces. The relatively high levels of As in rice straw are of concern in Bangladesh as this is the primary animal feed and can lead to arsenic transfer through the food chain. Levels of As in rice grain vary greatly and establishing a safe level is also complicated by (i) widely varying levels of rice intake in different countries, e.g. 450g dry wt/day in Bangladesh but <30 g dry wt/day in the USA, (ii) speciation of As in rice grain, which affects its toxicity and is predominantly inorganic (arsenite) in S. Asian rice, but predominantly dimethylarsinic acid (DMA) in USA rice [14] and (iii) the bioavailability of As in rice.

Our goal in this paper is to provide a perspective on:

1. The current status of As pollution of soils and groundwater used for irrigation water in Bangladesh.
2. Arsenic toxicity to rice and levels of As in rice straw and grain.
3. Safe levels of As in rice grain and soil from human safety and agricultural sustainability viewpoints.

2. Arsenic content of irrigation water and soils in Bangladesh

Surveys of As in irrigation water, soils, rice grain and rice straw were carried out between 2001-2005 to assess spatial variability in these parameters in individual tubewell command areas, in 5 upazilla (Bangladesh is divided into 481 upazilla, the smallest administrative unit) and nationwide.

2.1 National Scale: A preliminary geographically structured national scale survey of the As content of irrigation water and soils in Bangladesh involved collection of two samples from every other upazilla in rural agricultural areas for a theoretical total of 368 sampling sites. Where possible, samples of surface (pond) water and household tubewell water were also collected from points close to the irrigation tubewell. Surface water contained the lowest levels of As with 55 and 80 % of the samples containing <0.01 and 0.015 mg L^{-1}, respectively (Fig. 1a). Only one sample exceeded 0.05 mg L^{-1}. Seventy-seven percent of the irrigation STWs contained <0.1 mg L^{-1}, 15% contained between 0.1-0.2 mg L^{-1} and the remaining 8% contained >0.2 mg L^{-1} (Fig. 1a). The distribution of As in household well water was shifted to lower values than that in irrigation well water, with 37 and 79% of household well samples containing <0.01 and <0.05 mg L^{-1}, respectively (Fig. 1a).

The levels of As in irrigation and household STW's varied regionally with a spatial pattern similar to that reported earlier by the BGS-DPHE survey that had 3500 data points and therefore a much higher resolution [15]. Arsenic levels in STW's were much lower in the northern half than in the southern half of the country. The highest irrigation STW water As concentrations were found in the south-east and south-central areas below the confluence of the Ganges and Meghna rivers (Fig 1b). Fortunately for Bangladesh, much of the *boro* season rice grown in the highest groundwater As pocket to the SE of the Meghna river is irrigated with surface water so soil contamination with As is limited.

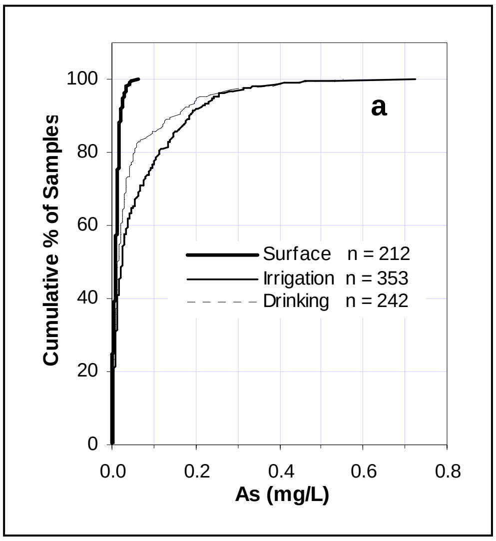
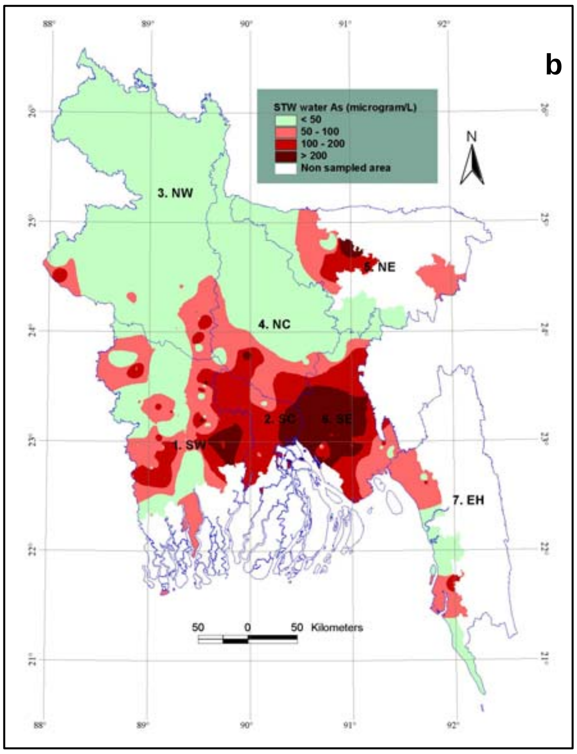


Fig. 1. a) Distribution of As concentrations in surface, household tubewell and irrigation tubewell water samples and b) Kriged map of As concentration in irrigation water from shallow tubewells



Surface soil (0-15 cm) As concentration was as high as 67.5 mg kg^{-1}, and also showed a geographic pattern. Like shallow tubewell irrigation water, soils in northern Bangladesh contained much less As than those in southern Bangladesh (Fig. 2a). Surface soils in the northern half of the country generally contained < 5 mg kg^{-1}, similar to background soil As levels for many regions of the world [16]. The highest surface soil As concentrations were found in the south-west quadrant, or more specifically the Ganges river basin, where much of the area contained >10 mg kg^{-1}. This distinct pattern indicates that the Ganges river sediments which created the current soil surface contained elevated levels of As, leading to higher background soil As levels. Detailed interpretation of this map

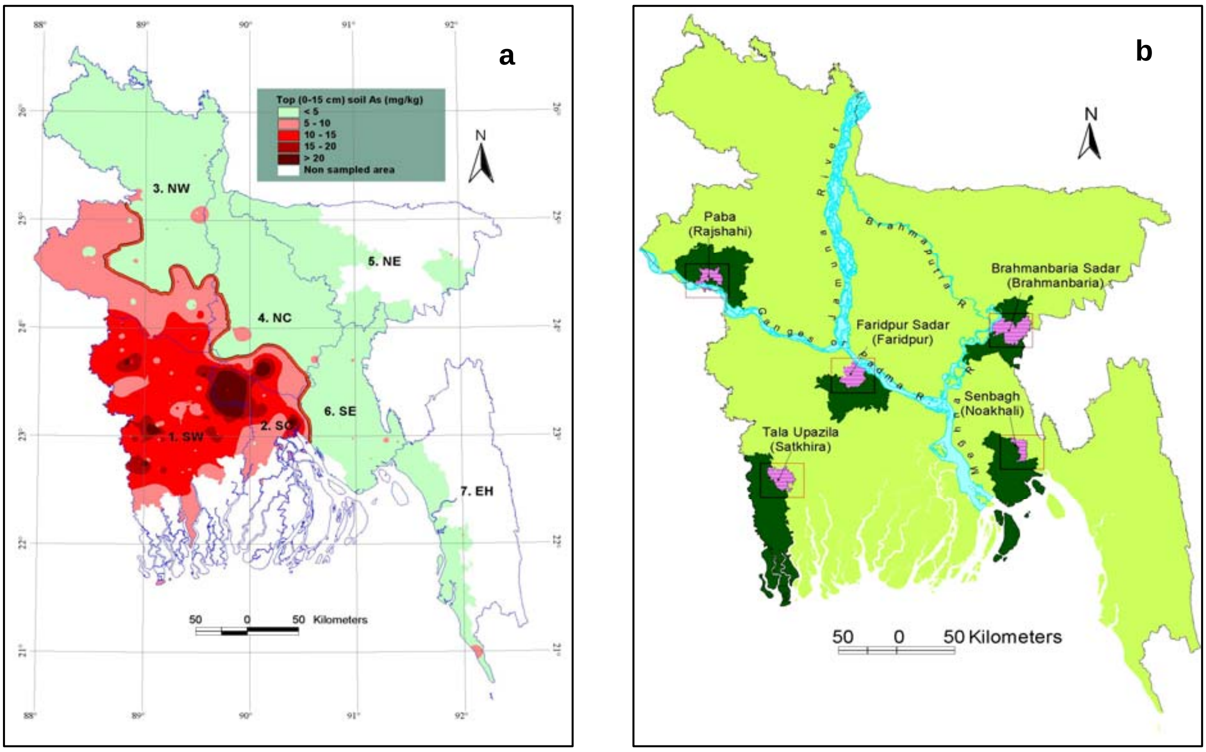


Fig. 2. a) Kriged map of As in surface soils (0-15 cm) of Bangladesh; solid line represents the 5 mg kg-1 boundary and b) Upazilla sampling sites and their associated districts

should not be attempted because of its low resolution, i.e. widely spaced sampling points, and more intensive sampling will undoubtedly identify contamination in areas shown as containing < 5 mg As kg^{-1} and greater spatial variability in the high As areas. Soil As concentrations in the south-west quadrant generally decreased with soil depth but were still between 5-15 mg kg^{-1} at the 30-45 cm depth, whereas sub-soil (15-30 and 30-45 cm) As levels were <5 mg kg^{-1} in the remainder of Bangladesh.

Inputs of As from irrigation water would add to the background soil As level. Twenty three percent of the irrigation well samples contained > 0.1 mg As L^{-1} which, assuming addition of 1 ha-m of irrigation water and retention of added As in soil, would increase soil As concentration in the top 15 cm of soil by 0.5 mg kg^{-1} per *boro* rice crop. Studies with natural soil columns containing low to high levels of As have shown high retention of As applied in irrigation water at levels of 1 and 2 mg As L^{-1} [17].

2.2 Upazilla Scale: More spatially intense sampling of irrigation water and soil was done in five upazilla (Fig. 2b) where a total of 455 samples were collected. Complex relationships between irrigation water and soil As levels were observed in this data set (Fig. 3). Of the 5 upazilla, only Paba upazilla in Rajshahi district with most irrigation water and soil samples containing <50µg As L^{-1} and <10 mg As kg^{-1}, respectively, could be considered relatively uncontaminated with arsenic. Soils in Brahmanbaria upazilla were similarly low in As but irrigation water levels were mostly >50µg As L^{-1} with a moderate number of samples containing >100µg As L^{-1}. Much of the *boro* rice area in this upazilla is under deep water during the summer monsoon season so that As added to soil by irrigation water can be remobilized and distributed laterally over large areas of land or even removed in rivers [5]. Irrigation water in Senbag upazilla was quite contaminated with As, with most samples containing >100µg As L^{-1}, yet the soil As content of all samples was <10 mg As kg^{-1}. This result suggests low retention of added As, which could partly be due to the lighter texture of soils in this region of Bangladesh and partly because of the relatively high P and low Fe concentrations in Senbag irrigation water, reducing the capacity to re-adsorb As as Fe in the water re-oxidizes. Indeed, the mean Fe:P mass ratio for irrigation water was 2.4 in Senbag compared to a range from 4.8-5.6 for the other upazillas. Soils in Tala and Faridpur upazillas were the most contaminated, with 91 and 85 % of samples containing >10 mg As kg^{-1} and 28 and 36 % containing >20 mg As kg^{-1}, respectively, which is consistent with the national survey results. Irrigation water in both upazillas was also widely contaminated with 68 and 41% of the samples from Tala and Faridpur, respectively, containing >100µg As L^{-1}. Consequently, agriculture in the central/south-west quadrant of Bangladesh is likely to be the most affected by arsenic.

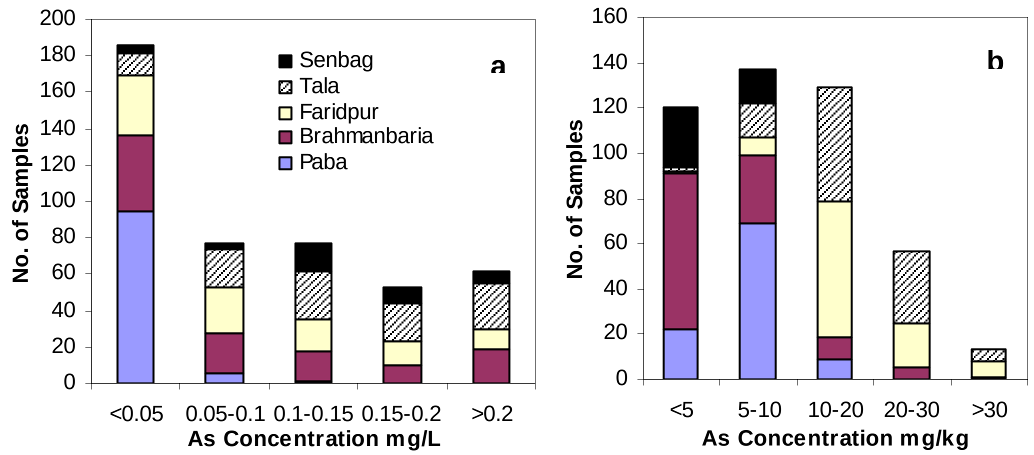


Fig. 3. Distribution of As concentrations in paired irrigation water (a) and soil (b) samples collected from five upazilla

An example of spatial pattern in soil As within an upazilla is given in Fig. 4. The spatial pattern is related to land type/land use, with the lower As areas being highland with limited use for boro rice production. In contrast, the higher As areas in the swath from from NW to SE are low lying *boro*-rice areas that are also cropped to rice in the summer monsoon season.

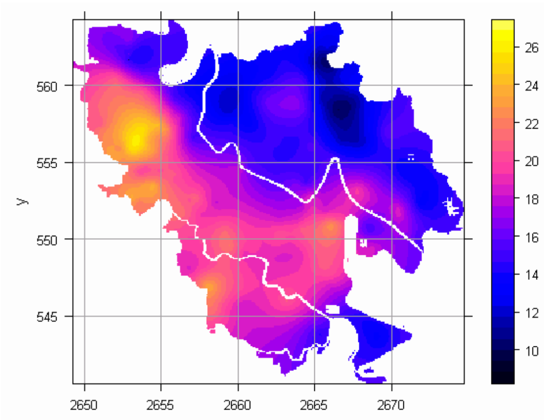


Fig.4. Kriged pattern of soil As in Tala Upazilla, Satkhira district

2.3 Tubewell Command Area Scale: Studies of individual command areas have shown spatially variable patterns of soil As [18, 33]. Such patterns are created as irrigation water is aerated during travel though irrigation channels and over fields. Aeration of the irrigation water oxidizes ferrous iron to ferric hydroxide which then precipitates and adsorbs both P and As from the water. Thus, both As and P are deposited on soil surfaces depending on the rate of oxidation of the irrigation water and the pathways of water flow over the command area (e.g. Fig. 5). Spatial pattern generated in this way can be permanent in fields that are cropped to rice during both the dry winter and the summer

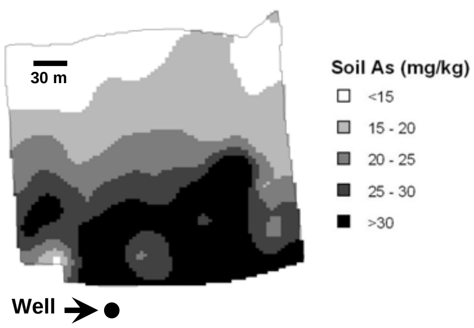


Fig.5. Kriged pattern of soil As in a command area of Poranpur village, Faridpur district [5]

monsoon seasons [5], or created in the *boro*-season but lost in the monsoon season for single cropped *boro* rice fields that are under deep water in the monsoon season [18]. The creation of spatial patterns in soil As is undesirable in that it can lead to sufficiently high levels of As to be phytotoxic to rice (see next section), it is impractical to measure soil As levels this intensively across all As affected areas in Bangladesh and it greatly complicates management of As in the soil environment.

3. Arsenic toxicity to rice and levels of As in rice straw and grain

3.1 Arsenic toxicity to rice: Several studies have shown that addition of fairly high levels of As, either directly to soil or in irrigation water, are phytotoxic to rice [19-21]. Our study at Poranpur is the only one that documents this occurring in a farmer setting where almost 20 yr of use of irrigation water containing 0.13 mg As L^{-1} created the soil As gradient shown in Fig. 5. At this site, yield of the variety BRRI dhan 29 declined progressively from 7-9 to 2-3 t ha^{-1} over a soil As gradient from 12 to 68 mg kg^{-1} in the two year study period [5]. The estimated yield reduction over the whole command area was 16%, which is a fairly substantial impact. Such a yield reduction over a significant portion of Bangladesh would not be acceptable to farmers or to policy makers, and the situation is likely to become worse over time with continued use of As contaminated irrigation water.

Strategies to address phytotoxicity to rice include identifying varietal tolerance and growing rice in more aerobic environments. Screening of varieties of rice grown in Bangladesh for tolerance to As has not been widely investigated, but our recent work indicates that there are some differences in varietal tolerance (Fig. 6a), and that all *boro*-season rice varieties released by the Bangladesh Rice Research Institute are vulnerable to high levels of available As [22]. Varietal tolerance to arsenic toxicity has been the goal of plant breeding programs in the southern USA for more than 30 years. Here, former cotton areas historically received large applications of both inorganic and organic arsenic pesticides [23]. Field screening of rice germplasm for tolerance to As has been done using monomethyl arsonic acid (MSMA). All USA rice cultivars (japonica sub-species) show some sensitivity to MSMA, whereas 25 of 125 Chinese lines were unaffected at the levels of MSMA used [24]. Of these, 24 were indica sub-species and 1 was a japonica sub-species. Arsenic contamination in Bangladesh is with inorganic As but there is evidence that MSMA is more toxic to rice than inorganic As species [25] so it is likely that the Chinese germplasm would be useful for Bangladesh.

Growing rice in a more aerobic environment on raised beds, where As is less available, has also shown potential to overcome As phytotoxicity to rice (Fig. 6 b) and [26]. This method of growing rice is not generally accepted and has had mixed results [27]. Our experience growing rice on raised beds in non-As affected areas of Bangladesh is that water inputs can be reduced by up to 40% while yields increase up to 30% with fewer plants in the field [28], so this approach is a viable strategy for As affected areas. In the USA and China, mid-season drainage of a paddies has also been used to reduce As availability and toxicity [29, 30] but is considered to have a yield penalty [24].

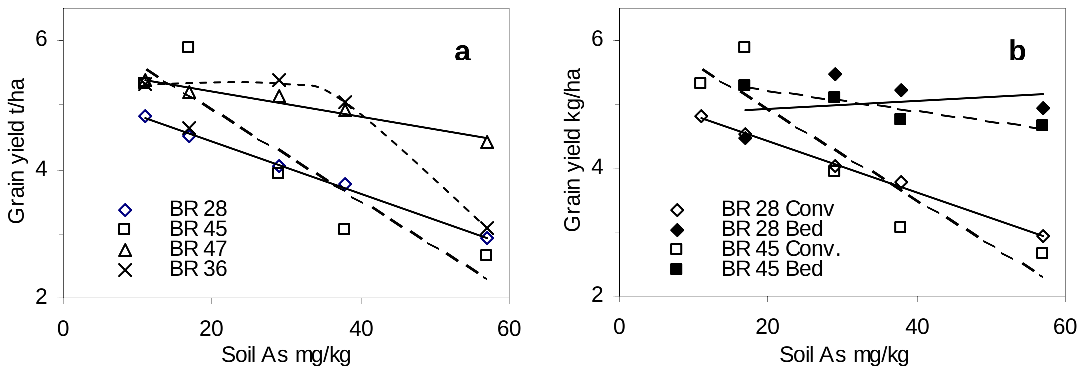


Fig. 6. Effect of increasing soil As content on rice grain yields at Poranpur village site, Faripur district for a) conventional paddy production of several BRRI rice varieties and b) conventional paddy and raised bed methods of rice production for two varieties

3.2 Arsenic content of rice straw and grain: The levels of As in rice straw and rice grain vary considerably. In general, the As concentration in straw is ~10x that in grain and levels in straw and grain are often positively correlated [17, 31] but the relationship my break down under conditions of As toxicity [5, 25]. Little is known about translocation of As from straw to grain and whether there are opportunities to reduce this. The comparatively high levels of As in rice straw are of concern as this is a primary animal feed in Bangladesh. Possible effects on animal productivity or health have not been investigated and there is potential for further human exposure to As via its movement through the feed/food chain.

We recently estimated a "normal" range for As in rice grain using 411 values obtained from all regions of the world [32]. Defining the "normal" range as between the 25^{th} to 75^{th} percentiles of a box plot of As concentration data gave a range from 0.08 to 0.20 mg kg^{-1}. Against this standard, much of the rice grain harvested from farmer fields in the national and upazilla surveys of Bangladesh would be classified as As contaminated as both mean and median values are above 0.20 mg kg^{-1} (Fig. 6). Rough separations between non-contaminated and contaminated sites based on As levels of < or > 50 µg L^{-1} for irrigation water and < or > 6 mg kg^{-1} for soil suggested that irrigation water As was more a driver of grain As levels than was soil As (Fig. 7).

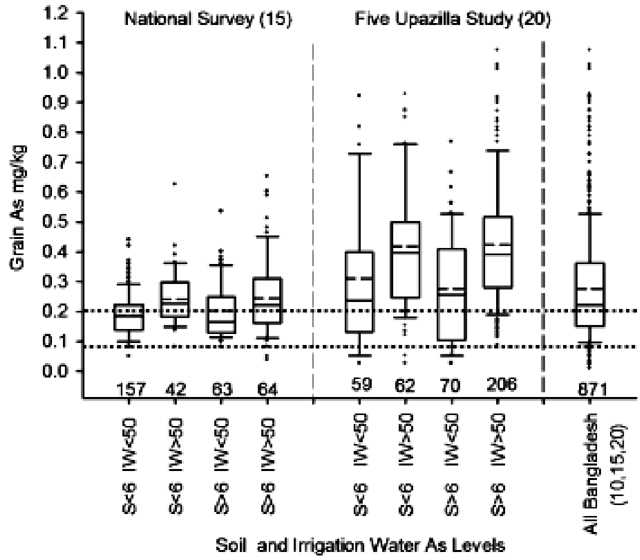


Fig. 7. Distribution of total As in rice grain samples from National and Upazilla surveys in Bangladesh. Horizontal dotted lines show the "normal" range for As in rice. Numbers above the *x*-axis are numbers of samples. The box represents data between the 25th and 75th percentiles. The whiskers (error bars) above and below the box indicate the 95th and 5th percentiles and dots above and below them represent outliers. Lines inside the box represent the mean (--) and median (-) values [32]

Several studies have reported good positive correlations between soil or irrigation water As and As in grain and straw of rice [17, 33, 34]. Our overall experience with large numbers of samples from farmer fields is that neither As in irrigation water or in soil are good predictors of As in rice grain. This lack of correlation is most probably because of variable water management practices and soil characteristics, and for the reasons given in the introduction section of this paper. In our study at the Poranpur site, grain but not straw As concentrations of variety BR 29 were reduced when phytotoxicity was severe (Fig. 8). It is possible that varietal differences in As concentrations in straw and grain may occur but this has not been reported and systematic studies under field conditions are needed.

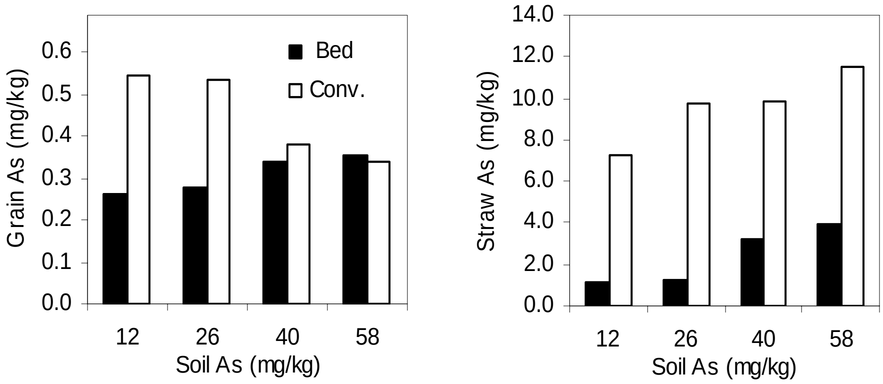


Fig 8. Effect of soil As levels on the As content of rice grain and straw for variety BR 29 grown under conventional paddy and raised bed management at Poranpur village site, Faridpur district [26]

4. Safe levels of As in soils and rice grain

4.1 Soil arsenic standards: Only China has a soil As standard for paddy soils, which is 30 mg kg^{-1} [35]. Several other countries have established soil arsenic standards that vary with land use or protection purpose. For example, Canada has recently (2001) established limits of 12 mg kg^{-1} based on human health risk for all land uses, but higher levels of 17 mg kg^{-1} for agricultural, residential and park use and 26 mg kg^{-1} for commercial and industrial uses from an environmental health perspective. Older (1983) standards include 10 mg kg^{-1} for food production in the UK and 20 mg kg^{-1} as good quality in Sweden. Remediation is required in Sweden at >50 mg kg^{-1}. An As level of ~200 mg kg^{-1} has been proposed for upland soils for protection of aquifers at a drinking water standard of 10 µg L^{-1} [36]. No similar soil As standard has been developed for flooded soils or based on risk of toxicity to rice. Critical soil As levels for yield reduction with different rice varieties have not been established. Available information suggests that even low levels of As may be problematic for Bangladesh rice varieties. Three (BR 28, BR 45, BR 47) of the four varieties shown in Fig 6a showed linear declines in grain yield with soil As levels above 10 mg kg^{-1}, while the fourth (BR 36) showed yield stability up to a soil As level of 30 mg kg^{-1}. Responses to soil As levels may differ amongst soil types and are strongly dependent on water management.

4.2 Safety of As in rice: China has a limit of 0.15 mg kg^{-1} for inorganic As in rice. The general limit for As in foods is 1 mg kg^{-1} in the UK and Australia, but these values are badly outdated. The provisional dietary intake level of 2.1 µg inorganic As kg^{-1} body weight established by WHO/FAO in 1989 allows for a daily intake of 126 µg inorganic As for a 60 kg person, which is well above the intake of 20 µg inorganic As that a person drinking 2L of water a day would receive at the current WHO standard of 10 µg inorganic As L^{-1}. In the absence of updated risk assessments for As in foods, the best strategy is to work from the WHO drinking water standard for As although the Bangladesh standard is set at 50 µg inorganic As L^{-1}. The reason for the focus on inorganic As is that organic As forms are widely considered to be much less toxic than the inorganic forms. The forms of As in rice grain as well as their bioavailability have only recently begun to be studied. Our compilation of all modern (post 1996) As speciation data for rice grain suggested that there are two types of rice [37]. At low concentrations of grain As, both rice types contain predominantly inorganic As but as As concentrations increase the form of As becomes predominantly dimethyl arsinic acid (DMA) in one type while remaining predominantly inorganic As in the other type (Fig. 9). In this study, rice from the USA, Australia and China was mostly found to be the DMA type, while rice from Europe and S. Asia was the inorganic As type.

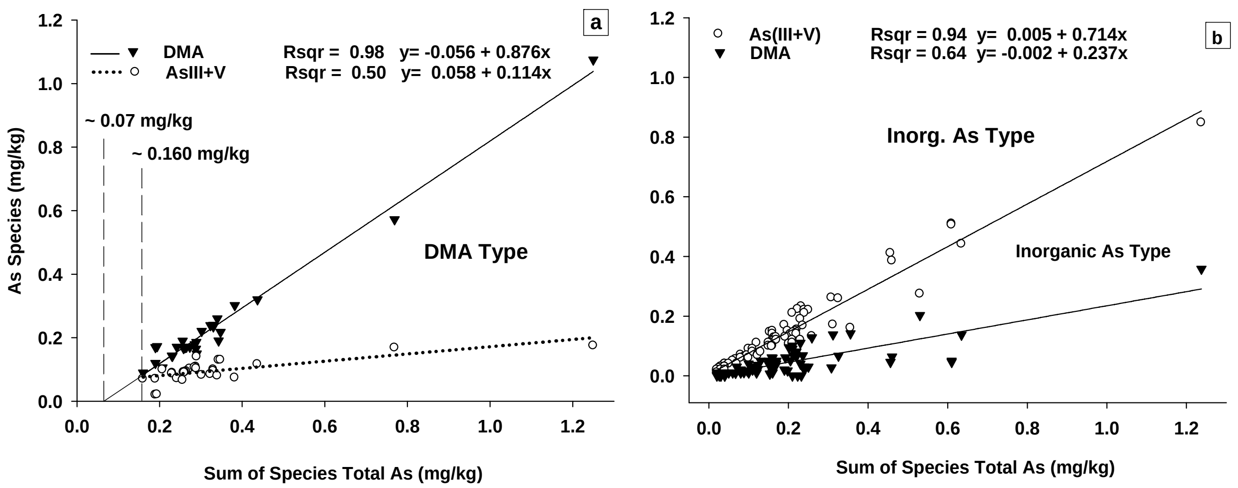


Fig. 9. Categorization of As speciation data for rice into DMA and Inorganic As rice types [37]

Although still somewhat controversial, the human toxicity of DMA is thought to be less than that of inorganic As [37], so that it may be appropriate to base risk assessment on the inorganic As content of rice. In this case, the DMA rice type would be much more desirable as it has less inorganic As than the inorganic As rice type when total As exceeds 0.09 mg kg^{-1}. The only study on bioavailability of As in rice [38] found that inorganic As in rice was assimilated much more readily than DMA, also supporting a focus on inorganic As. The safety of rice grain can therefore be evaluated by comparisons with standards for inorganic As in drinking water. For Bangladesh, rice grain generally contains 80% inorganic As [39, 40] and the average adult daily intake of rice is 450g dry wt. The water equivalent standards for total As in rice using these figures and assuming 85% bioavailability of inorganic As in rice

Table 1. Equivalent standards for As in drinking water and rice for 450g (DW) rice and two levels of water intake

Water As Standard µg L^{-1}	Daily As Intake from water - µg 2L	4L	Equivalent Rice Total As concentration* - mg kg^{-1} 2L	4L
10	20	40	0.065	0.130
50	100	200	0.330	0.650

*values rounded to nearest 0.005 mg kg^{-1}

[38] are shown in Table 1. By comparing the data in Table 1 with values for As in Bangladesh rice (Fig. 7), it can be seen that almost none of the rice produced in Bangladesh would meet a standard equivalent to the WHO limit for As in drinking water of 10 µg L^{-1}.   In contrast, 67 and 93% of Bangladesh rice samples would be below a rice equivalent value based on the Bangladesh drinking water standard of 50 µg L^{-1} for daily per capita water intakes of 2 and 4 L, respectively. Overall, it is clear that As intake from both water and rice needs to be included when considering human health based standards for As in Bangladesh, and also for other Asian countries with high rice consumption rates.

Unfortunately, there is emerging evidence that tolerance of rice to As is not related to exclusion of As from rice straw or grain and tolerant varieties can contain very high levels of As in both straw and grain [8,22]. Simply breeding rice for As tolerance will likely increase human exposure to As from rice. In this case, adoption of water management strategies to reduce As in rice, and a better understanding of the speciation and relative toxicities of the different As species in rice grain and straw, thus becomes of paramount importance for protecting human and animal health.

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