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RESEARCH ARTICLE

Arsenic budgeting in a long-term irrigation-contaminated rice field of the Bengal Delta Plain

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Abstract

Arsenic (As) contamination of groundwater-irrigated rice agroecosystems presents a major environmental and public health challenge across the Bengal Delta Plain (BDP). Although long-term irrigation is thought of as a dominant As input pathway, there is a lack of studies that have quantitatively reconciled field-scale As influx with measured soil accumulation. This study establishes a 33-year As mass balance for a rice field in Nadia district, West Bengal, integrating measured irrigation-water As concentrations, rice grain and straw As contents, crop yields and topsoil As levels. Annual groundwater irrigation introduced 207 mg As m^{-2} yr^{-1}, whereas biomass removed only 3.66 mg As m^{-2} yr^{-1} (< 2 % of inputs). The resulting net accumulation of 203.4 mg m^{-2} yr^{-1} predicted an enrichment of 39.9 mg kg^{-1} in the 0-15 cm soil layer, aligning closely with the measured value of 37.9 mg kg^{-1}. This agreement indicates minimal leaching or lateral losses and reflects strong Fe-mediated retention under flooded redox conditions. The findings confirm that groundwater irrigation alone accounts for long-term As build-up in BDP paddy soils and provide a quantitative basis for anticipating future soil burdens under continued reliance on As-bearing aquifers.

Keywords: accumulation; groundwater irrigation; ICP-MS; iron redox cycling; mass balance; paddy

Introduction

Arsenic (As) contamination in agroecosystems has emerged as a major global environmental and public health concern, primarily because of its chronic toxicity and efficient transfer through water and food systems. Long-term exposure to As is linked to skin lesions, cardiovascular disorders, diabetes, neurological impairment and various cancers (1). An estimated 300-500 million people worldwide are exposed to As concentrations exceeding the World Health Organisation (WHO) drinking-water guideline and about 1.1 % of global croplands are contaminated with As, with the highest risks concentrated in South and Southeast Asia (2, 3). In addition to drinking water, dietary intake has become a dominant exposure pathway in regions where rice constitutes the staple food (4). Rice is particularly prone to As accumulation due to its cultivation in flooded soils (5). Under anaerobic conditions, Fe (III) oxyhydroxides dissolve, releasing sorbed As into soil pore-water. This promotes the formation and uptake of arsenite (As (III)), a species that is more mobile and more easily transported across rice root membranes via silicon (Lsi1, Lsi2) and phosphate (OsPT2, OsPT8) transport pathways. Numerous studies from Bangladesh, India, China and Vietnam have reported elevated As concentrations in rice grain when irrigated with As-bearing groundwater (6). Given rice's central contribution to daily caloric intake in these regions, even moderate increases in grain As substantially elevate dietary exposure risks, particularly for vulnerable populations-rural households, children and women.

The Bengal Delta Plain (BDP) represents the most severely affected As hotspot globally (7). Here, shallow alluvial aquifers are naturally enriched with geogenic As and are extensively exploited for irrigation. Groundwater As concentration of BDP frequently exceeds 300 µg L^{-1} and, in extreme cases, it may reach up to 4600 µg L^{-1} (8). Continuous use of such groundwater has delivered substantial As loads to paddy soils. Long-term application has produced measurable increases in soil As, although the magnitude of enrichment varies widely due to differences in sediment lithology, aquifer geochemistry, redox dynamics, organic carbon availability, seasonal flooding regimes and irrigation intensity (9). Importantly, the BDP displays marked spatial heterogeneity in total soil As concentrations, with values reported in the range of <5 to 95.3 mg kg^{-1} across West Bengal and Bangladesh (10-12). This variability reflects contrasting aquifer geochemistry, sediment age, irrigation intensity and hydropedological settings, which together create a patchwork of low-, moderate- and high-risk As zones across the delta. West Bengal, forming the western segment of the BDP, ranks among India's highest rice-producing states, much of which is cultivated on As-impacted soils (13).

A balance sheet or mass-balance approach provides a means to quantify whether the observed soil As concentrations of paddy fields reflect the cumulative net input of As over time. This involves comparing the annual influx of As through irrigation water with the annual outflux via harvested biomass export. If inflow exceeds outflow, As accumulates in soil; if outflow equals or exceeds inflow, soil As remains stable or declines. Importantly, this approach distinguishes whether soil As levels are primarily driven by irrigation inputs, internal transformation and retention processes, or additional external sources. Although it is widely assumed-both scientifically and in policy discourse-that groundwater irrigation is the dominant source of As enrichment in BDP paddy soils, this assumption has rarely been validated through quantitative mass-balance evaluations. No prior study has explicitly demonstrated, using long-term field-scale data, that irrigation-derived As alone can account for the measured soil As burden. The objectives of this study are to quantify annual As influx from irrigation, estimate outflux through grain and straw removal, determine net annual As retention and evaluate whether measured soil As aligns with modelled cumulative enrichment over 33 years. By establishing this direct correspondence between modelled and measured soil As levels, this study provides the first field-based proof-of-concept that irrigation water is the principal contributor to long-term As accumulation in BDP paddy soils. We hypothesise that the soil acts as a net sink for As and that long-term groundwater irrigation has progressively enriched the plough layer. Establishing this balance is essential for predicting future soil As burdens and informing sustainable irrigation management in As-affected rice agroecosystems.

Materials and Methods

Composite surface soil samples (0-15 cm) were collected in April, immediately after the harvest of *Boro* rice, from 3 locations within a 6.2 m × 3.3 m plot identified in a rice-growing farmer's field at Mitrapur village (22.9954 °N, 88.6125 °E; elevation 9 m), situated in the alluvial tract of the BDP in Nadia district, West Bengal, India. (Fig. 1). The field has been under a consistent *Aman* rice-*Boro* rice-fallow rotation since 1988. The irrigation, tillage and fertiliser practices remained more or less consistent over the years. While *Aman* rice cultivation relied primarily on monsoon rainfall with occasional groundwater support, *Boro* rice cultivation during the dry season depended entirely on groundwater irrigation.

After air-drying and passing through a 2 mm sieve, the soil samples were pooled, processed and analysed for total As using microwave-assisted digestion and ICP-MS (PerkinElmer NexION 300) (7). The processed soil samples were also analysed for pH, electrical conductivity (EC), texture, total organic carbon (TOC), free $CaCO_3$ and amorphous as well as crystalline Fe- and Al-oxides, following standard procedures described (14, 15). All samples were analysed in triplicate. Recently harvested grain and straw samples were also collected in triplicate from the farmer of the studied field for analysing As contents (7). Irrigation water samples were collected in triplicate from the groundwater-fed pump supplying the field. Water samples were filtered (0.45 µm), acidified, stored in a refrigerator (4 °C) and analysed for As concentration using ICP-MS (16).

The balance sheet approach computed As influx through irrigation water and outflux via harvested rice grain and straw. Annual irrigation water input, average grain yield and grain-to-straw ratio data were collected from the farmer and matched with previous literature (17, 18). A reference soil was additionally sampled from an uncultivated site at Mandouri farm (Bidhan Chandra Krishi Viswavidyalaya, Mohanpur, Nadia district), which has no known history of anthropogenic As contamination. This

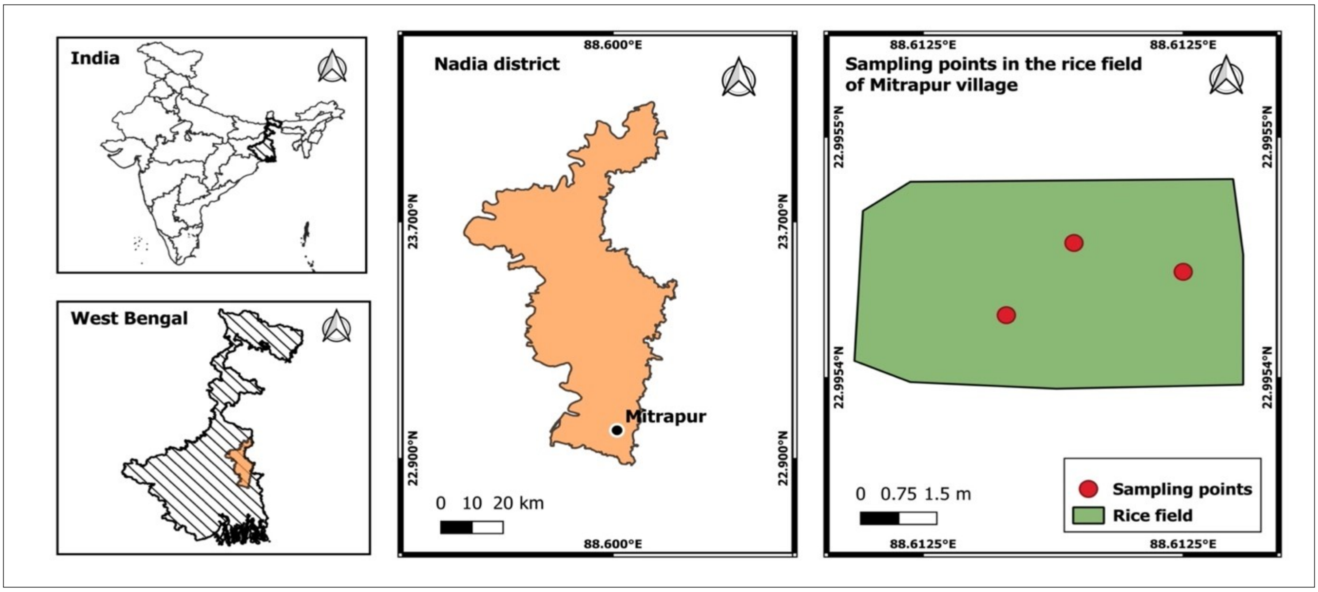

Fig. 1. Location of the studied rice field in Mitrapur village, Nadia district, West Bengal, India, within the Bengal Delta Plain (BDP) (The map has been generated using QGIS 3.40).

sample was analysed in triplicate to determine the background As concentration of the studied region.

Results and Discussion

Physico-chemical properties of the experimental soil, irrigation water and reference soil

The Mitrapur soil exhibited a slightly alkaline pH and was texturally classified as sandy clay with a notably high clay fraction, substantial total organic carbon and appreciable free $CaCO_3$ content. Total As concentrations in the irrigation water, Mitrapur soil and the reference Mandouri soil were 142 μg L^{-1}, 37.9 mg kg^{-1} and 7.91 mg kg^{-1} respectively. The As level in the Mitrapur soil fits well within the documented spatial range reported for the western BDP (<5-95.3 mg kg^{-1}) (10-12), while the reference soil corresponds to the global background (5-7.5 mg kg^{-1}) (19). The experimental soil was also rich in multiple potential As-adsorbing phases, including both amorphous and crystalline Fe and Al oxides, along with abundant clay minerals, $CaCO_3$ and organic matter- attributes that hold particular relevance for interpreting As retention behaviour in later sections of this study. These sorbents promote As partitioning to the solid phase over pore-water dissolution, immediately limiting bioavailability and toxicity risks during flooding. Long-term, they sustain As retention despite redox fluctuations, explaining low pore-water As despite high soil totals in BDP paddies. A complete summary of the physicochemical properties of both soils and the irrigation water is provided in Table 1.

Table 1. Physico-chemical properties of the experimental soil, irrigation water and reference soil

Properties	Values
Experimental (Mitrapur) soil	
pH	7.67 ± 0.05
Electrical Conductivity (dS m^{-1})	0.86 ± 0.02
Texture	Sandy clay
Clay %	47.9 ± 1.6
Total organic carbon (g kg^{-1})	17.2 ± 0.8
Free calcium carbonate (%)	4.9 ± 0.3
Total As (mg kg^{-1})	37.9 ± 0.9
Amorphous Fe (%)	2.16 ± 0.12
Amorphous Al (%)	0.93 ± 0.05
Crystalline Fe (%)	1.02 ± 0.07
Crystalline Al (%)	0.66 ± 0.05
Irrigation water sample of Mitrapur	
pH	7.19 ± 0.04
Electrical Conductivity (dS m^{-1})	0.05 ± 0.01
As (μg L^{-1})	142 ± 8
Reference (Mandouri) soil	
Background As (mg kg^{-1})	7.91 ± 0.42

Influx of arsenic to the rice field through irrigation water

The annual As influx to the studied rice field was determined from the total volume of irrigation water applied and its measured As concentration. Based on field-level information obtained from the farmer and validated against regional irrigation estimates for *Boro* season paddy cultivation, the annual irrigation water application in the studied rice field was approximately 1458 mm, equivalent to 1458 L m^{-2} yr^{-1} (17). This estimate aligns closely with reported irrigation inputs (1200-1500 mm) for *Boro* rice in the groundwater-irrigated command areas of West Bengal, where extended irrigation periods are required due to limited monsoonal recharge (20, 21). The substantial irrigation demand reflects both the water-intensive nature of rice cultivation and the dependence on As-bearing shallow aquifers for maintaining flooded field conditions throughout the growing season. The irrigation groundwater contained 142 μg L^{-1} of dissolved As, resulting in an annual As load of 207 mg m^{-2} yr^{-1} entering the soil system (Table 2). This magnitude of As influx is consistent with long-term irrigation practices widely reported across the BDP, where shallow tubewells serve as the primary water source for *Boro* rice cultivation. In similar agroecological settings of BDP, groundwater irrigation has been shown to introduce between 50 and 450 mg As m^{-2} yr^{-1}, depending on tube well depth, aquifer geochemistry and irrigation intensity (18, 20, 22, 23). The current estimate falls within this regional range, emphasising that the studied site reflects a typical As-loading scenario rather than an isolated case of contamination. The chemical form of As in irrigation water strongly governs its behaviour in paddy soils. Groundwater in the BDP typically contains As released through reductive dissolution of Fe (III) oxyhydroxides and when applied to flooded fields, rapidly encounters reducing soil conditions that enhance As mobility. Puddling further restricts percolation, causing most irrigation-derived As to accumulate in the plough layer, where studies consistently report steep vertical gradients with surface enrichment (22, 24). Seasonal redox oscillations promote recurrent Fe-As release and re-sequestration cycles, making the 207 mg m^{-2} yr^{-1} influx a persistent and geochemically active input shaping long-term soil As accumulation.

Outflux of arsenic from the rice field

The primary pathway through which As leaves flooded rice fields is via export of harvested biomass, particularly the grain and straw fractions. Based on field-level information obtained from the farmer and consistent with the reported average grain yield of 2573 kg ha^{-1} yr^{-1} for rice cultivated in the BDP, the productivity of the studied field closely matches regional yield benchmarks for long-term groundwater-irrigated *Boro-Aman* rice systems (18). Given this yield level and the measured grain-As concentration of 0.382 mg kg^{-1}, the estimated annual As removal through harvested rice grain is relatively modest, indicating that only a small fraction of the total As input is exported via edible biomass. This is consistent with the understanding that rice grain, despite being the main dietary exposure vector for humans, accumulates lower As concentrations relative to vegetative tissues (7, 25). In contrast, rice straw exhibited a substantially higher average As concentration (6.01 mg kg^{-1}), though produced in comparable biomass (0.288 kg m^{-2} yr^{-1}). The much higher straw As concentration reflects the strong tendency of inorganic arsenite to bind to thiol-rich cell walls and iron plaque on root and stem tissues (26, 27). However, although straw, As concentration is high, its contribution to net As outflux depends critically on post-harvest residue management. In this field, straw is traditionally removed from the site for livestock feed and household fuel, thus constituting a significant As export pathway. If straw were retained or incorporated, most of this As would remain in the soil system, adding to the long-term accumulation. Together, grain and straw export removed only 1.83 mg As m^{-2} per cropping season, or 3.66 mg As m^{-2} yr^{-1} for 2 rice crops annually (Table 2). This value is 2 orders of magnitude lower than the annual As input through

Table 2. Balance sheet of arsenic in the experimental rice field

Influx to Rice Field		
Parameters	Values	Units
Irrigation water inflow per square meter	1458	mm
Volume of water in L	1458	L
Arsenic concentration in irrigation water	0.14	mg L^{-1}
Total annual arsenic influx to the paddy field through irrigation water	**207.0**	**mg m^{-2} year^{-1}**
Outflux from Rice Field		
Average rice grain productivity yr^{-1}	2573	kg ha^{-1}
Average rice grain productivity yr^{-1}	0.26	kg m^{-2}
Average rice straw productivity yr^{-1}	2880	kg ha^{-1}
Average rice straw productivity yr^{-1}	0.29	kg m^{-2}
Measured arsenic concentration in rice grain	0.38	mg kg^{-1}
Measured arsenic concentration in paddy straw	6.01	mg kg^{-1}
Total amount of arsenic leaving the paddy field through agricultural products in each season	1.83	mg m^{-2} season^{-1}
Total amount of As leaving the paddy field through agricultural products annually	**3.66**	**mg m^{-2} year^{-1}**
Mass Balance in Rice Field		
Arsenic accumulation in Paddy soils (Inflow through irrigation water - Outflow through agricultural products) or arsenic excess in soil	203.4	mg m^{-2} year^{-1}
Volume of the top 15 cm of soil m^{-2}	0.15	m^{3}
Soil density	1.4	g cm^{-3}
Soil density	1400	kg m^{-3}
Mass of the top 15 cm of soil m^{-2}	210	kg
Increase in soil arsenic content after distribution of excess arsenic in the top 15 cm of soil	0.97	mg kg^{-1} year^{-1}
Increase in soil As content due to the excess amount of As supplied in the last 33 year	31.96	mg kg^{-1}
Background arsenic concentration in soil	7.91	mg kg^{-1}
Total calculated arsenic concentration in soil over a period of 33 year	**39.9**	**mg kg^{-1}**
Actual concentration of As in soil	37.9	mg kg^{-1}

irrigation (207 mg m^{-2} yr^{-1}). Only a very small fraction, typically <1-3 % of incoming As is removed in crop biomass, while the remainder accumulates in soil or undergoes secondary redistribution (28-31). Thus, the agricultural output stream is a minor loss pathway relative to irrigation-driven input.

Net arsenic accumulation and soil mass balance

Subtracting the annual outflux of As through harvested biomass (3.66 mg m^{-2} yr^{-1}) from the annual influx via irrigation water (207 mg m^{-2} yr^{-1}) indicates a net As accumulation rate of approximately 203.4 mg m^{-2} yr^{-1} in the soil system. When this excess As is distributed across the upper 0.15 m of the plough layer, with an estimated soil mass of 210 kg m^{-2}, the corresponding increase in soil As concentration is calculated to be around 0.97 mg kg^{-1} yr^{-1}. The assumed bulk density of 1.4 Mg m^{-3} for the surface paddy soil is consistent with values reported for fine-textured, long-term puddled rice soils of the BDP, which typically range between 1.3 and 1.5 Mg m^{-3}due to repeated wetting-drying cycles, compaction from tillage and high clay and organic matter contents (32, 33). This estimation thus provides a realistic basis for assessing As enrichment in the surface horizon of the studied irrigated rice field. Over the 33-year cultivation period, the estimated cumulative enrichment of As in the studied paddy soil was approximately 31.96 mg kg^{-1} (Table 1). When added to the measured local background concentration of 7.91 mg kg^{-1}, the predicted total As concentration in the topsoil is 39.9 mg kg^{-1}. This value corresponds remarkably well with the actual measured concentration of 37.9 mg kg^{-1}, differing by only 2.0 mg kg^{-1}, thereby validating the balance-sheet approach. Such close agreement implies that, As introduced through irrigation has largely remained within the plough layer, with minimal vertical or lateral redistribution.

The negligible discrepancy between the modelled and measured values indicates that leaching losses were minimal, owing to the presence of abundant sorptive phases in the studied soil that are well known to immobilise As under flooded-field redox cycles. The strong capacity of BDP paddy soils to retain As is well established (15, 34). Both amorphous and crystalline Fe (III) (oxyhydr) oxides act as the principal sinks, binding As (V) and As (III) through stable inner-sphere complexes that can persist despite partial reductive dissolution (15). Aluminium oxides provide additional high-affinity sorption sites, especially for As (V) under near -neutral pH conditions (35). High clay contents further enhance As retention via ligand-exchange reactions on smectite and mica edge surfaces (36). Calcium carbonate promotes Ca-bridged arsenate associations, while soil organic matter stabilises Fe-As coprecipitates during repeated redox fluctuations (37, 38). Together, these interacting mechanisms markedly limit downward As transport, explaining the minimal leaching losses observed.

Surface runoff is also minimised due to bunded field hydrology, limiting lateral export of As-enriched water. Gaseous or microbial volatilisation is likely to be minor as well, because As-methylating microbial communities in submerged soils exhibit sporadic activity and typically produce only trace quantities of volatile As species (39). Collectively, these findings affirm that long-term irrigation with As-contaminated groundwater has been the dominant and persistent source of As accumulation in this rice soil. Furthermore, the strong retention within the root zone underscores the critical role of internal soil processes-particularly Fe-mediated sorption, coprecipitation and redox buffering-in governing As residence and mobility in intensively flooded agroecosystems of the BDP. These validated mass-balance results provide a quantitative basis for future As modelling by demonstrating the overwhelming dominance of irrigation inputs (≈99 % of net As loading) and the high retention efficiency of the soil when data are collected over long time scales in targeted fields. They further provide an evidence-based basis for endorsing mitigation options such as alternate wetting-drying, upland rice cultivation and Fe-enhancing amendments to minimise prolonged flooding and curb long-term soil As accumulation, thereby strengthening their policy relevance.

Conclusion

The 33-year mass balance assessment demonstrates that As accumulation in the studied BDP rice soil is overwhelmingly controlled by sustained inputs from As-contaminated groundwater irrigation. Spanning more than 3 decades, this long-term assessment captures cumulative irrigation-driven As loading under consistent rice cultivation, offering a robust temporal framework rarely available for soil As studies in intensively managed agroecosystems. Although the origin of groundwater As is geogenic, the resulting soil pollution is anthropogenically driven through continuous irrigation inputs. Annual As influx via irrigation exceeded outflux through harvested biomass by nearly 2 orders of magnitude, resulting in a substantial net retention. The predicted cumulative enrichment in the plough layer closely matched the analytically measured concentration, confirming that As has remained largely confined to the topsoil with negligible vertical leaching or lateral loss. This strong retention reflects the dominance of Fe-mediated sorption, coprecipitation and redox-driven cycling in flooded rice soils. The findings conclusively demonstrate that long-term groundwater irrigation alone is sufficient to account for present-day soil As burdens in the BDP. This underscores the need for urgent policy and agronomic interventions, including improved irrigation water management, safe aquifer sourcing, deficit or alternate wetting–drying irrigation, judicious cultivar selection with low As accumulation potential and other mitigation strategies to limit further soil loading and reduce future dietary exposure risks. Future research should prioritise field-scale evaluation of alternative irrigation regimes (e.g., intermittent flooding or low-As surface water blending) coupled with soil-plant As mass balance modelling to quantify their effectiveness in slowing or reversing As accumulation in long-term irrigated rice systems of the BDP.

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Authors' contributions

AR conceived the study, designed the methodology, carried out the investigation, sampling, formal analysis and prepared the original manuscript draft. SPD supervised the work and contributed to funding acquisition, conceptualisation, methodology development and provision of resources. KMM provided supervision, contributed to methodology and assisted in funding acquisition. MB supported conceptualisation, project administration and contributed to writing. PR provided resources and participated in review and editing. DG performed validation and data curation. DR carried out data curation and visualisation. V conducted formal analysis and contributed to review and editing. MS participated in the investigation. DD performed validation. SM handled formal analysis and writing, review and editing. PK contributed to visualisation writing, review and editing. All authors read and approved the final manuscript.

Compliance with ethical standards

Conflict of interest: Authors do not have any conflict of interests to declare.

Ethical issues: None

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