

A Review on Sources, Hydrochemical and Temporal Changes of Arsenic Contamination in the Groundwater of Ballia and Ghazipur Districts

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ABSTRACT

Arsenic contamination in groundwater has emerged as a significant environmental and public health challenge, both nationally and globally. Recent assessments of groundwater samples from key regions in the northeastern states of India reveal arsenic concentrations ranging from 50 to 986 µg/L, exceeding the permissible limits set by the World Health Organization (WHO) and the Bureau of Indian Standards (BIS), which are 10 µg/L and 50 µg/L, respectively. The seasonal behavior of arsenic is largely governed by geochemical processes, wherein iron(III) oxyhydroxide decomposition during the monsoon season releases arsenic into the groundwater, while its adsorption onto iron(III) oxyhydroxide occurs during the pre-monsoon season.

This study examines the sources, hydrochemical characteristics, and seasonal variations of arsenic in groundwater within the Ballia and Ghazipur districts of Eastern Uttar Pradesh. As predicaments in the environment is due to its mobilization under natural geogenic conditions as well as anthropogenic activities. Arsenic mineral is not present in As contaminated alluvial aquifer but As occurs adsorbed on hydrated ferric oxide (HFO) generally coat clastic grains derived from Himalayan mountains. As is released to the groundwater mainly by bio-remediated reductive dissolution of HFO with corresponding oxidation of organic matter. The development of strongly reductive dissolution of mineral oxides (Fe and Mn) at near-neutral pH may lead to desorption and ultimately release of As into the groundwater. As release through geochemical process is more important factor in alluvial aquifers causing As contamination rather than sources of arsenic. During the monsoon, rising water tables and flood conditions contribute to arsenic dilution, whereas prolonged moisture retention restricts groundwater movement, affecting arsenic dispersion. This study highlights the impact of hydrological cycles on arsenic behavior, providing insights for effective groundwater management in arsenic-affected regions.

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Keywords: Arsenic contamination, groundwater hydrochemistry, seasonal variation, redox potential, monsoon impact

INTRODUCTION

India, a nation characterized by its vast population density, is facing an acute crisis of potable water due to the imbalance between its land area and population. Despite covering only 2.4% of the Earth's surface, India supports approximately 17% of the global population, resulting in severe water scarcity challenges (Das, D., Chatterjee et al., 1995; Graeme KA et al., 1998; Singh AK, 2004; Mukherjee A et al., 2006; Ding, A., Yang, S. et al., 2007). Historically, the contamination of surface water by sewage and industrial waste has rendered it unsuitable for human consumption and industrial applications. Consequently, both urban and rural populations are increasingly dependent on groundwater and rainwater for their water needs. However, the uneven distribution of groundwater across the country exacerbates the crisis.

India faces a dual threat of water pollution—both naturally occurring and anthropogenic—posing significant concerns regarding the quality of drinking water. Among these concerns, arsenic contamination in groundwater is particularly alarming and affecting greater population in India (Mishra et al., 2024). Arsenic, a potent carcinogen, has been the subject of extensive research to understand its origin and mobility within groundwater systems. The first documented case of arsenic poisoning in the Indian subcontinent dates back to 1983, although arsenic contamination in West Bengal's groundwater was first reported in 1978 (Khan HR, 1994; Milliman JD et al., 1995; Bhattacharya P et al., 1999). Since then, widespread occurrences of naturally occurring arsenic have been identified across the Ganges river plains, Uttar Pradesh, northeastern states, and other regions of India (Mishra et al., 2023). Through this references add more sentence and then link with arsenic contamination in Indo-Gangetic plain

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Mishra, R., Datta, S.P., Golui, D., Meena, M.C., Dwivedi, B.S., Rahman, M.M., Bandyopadhyay, K., Bhatia, A. and Pandey, P.S., 2023. Evaluation of different extractants to estimate bioavailable arsenic in soil. *Communications in Soil Science and Plant Analysis*, 54(17), pp.2459-2474.

Mishra, R., Datta, S.P., Golui, D., Meena, M.C., Raza, M.B., Rahman, M.M., Chaudhary, M., Behera, S.K., Chaudhary, A., Upadhyay, D.P. and Shukla, A.K., 2024. Mapping arsenic contamination and health risk assessment of arsenic in agricultural soils of eastern India. *Water, Air, & Soil Pollution*, 235(9), p.559.

ARSENIC IN THE ENVIRONMENT: OCCURRENCE AND TOXICITY

Arsenic (As) is rarely found in its free elemental form and primarily occurs in combination with sulfur, oxygen, and iron. It exists in various oxidation states—As(V), As(III), As(0), and As(−3)—with As(III) being 60 times more toxic than As(V) (Mabuchi et al., 1979). In aquatic environments such as ponds, seas, and lakes, arsenic undergoes biomethylation, leading to the presence of arsenite, arsenate, mono-methylarsonate (MMA), and di-methylarsinate (DMA). The inorganic forms of arsenic are about 100 times more toxic than the methylated variants (Thompson, 1993), and As(III) is considered more hazardous than As(V) (NAS, 1977). The ratio of As(III) to As(V) varies widely based on redox conditions in geological environments (Braman and Foreback, 1973; Andreae, 1977; Shaikh and Tallman, 1978; Ying et al., 2011; Molinari et al., 2013; Kanoua and Merkel, 2017).

Human exposure to arsenic occurs through multiple pathways, including air, food, water, and soil (Cullen and Reimer, 1989; NRC, 1999; Khan et al., 2020; Mishra et al., 2024). Arsenic contamination of groundwater is a significant threat to public health and ecosystems worldwide. Millions of people are exposed to high levels of arsenic, not only through drinking water but also via food consumption, making it a global concern (Bundschuh et al., 2010). Arsenic is widely distributed in nature, constituting about 0.00005% of the Earth's crust and ranking 12th in abundance in the human body (Mandal and Suzuki, 2002). The estimated global average concentration of arsenic is 5 mg/kg in soil, 1–2 µg/L in open seawater, and below 10 µg/L in unpolluted surface and groundwater.

GEOCHEMICAL AND ENVIRONMENTAL FACTORS INFLUENCING ARSENIC CONTAMINATION

The presence of arsenic in natural water systems is largely governed by geochemical conditions, including basin-fill deposits of alluvial-lacustrine origin, volcanic formations, geothermal inputs, mining activities, and landfill leachates (Welch et al., 1988; Korte and Fernando, 1991). Local geological, hydrological, and geochemical factors significantly influence arsenic mobilization in groundwater (Bhattacharya et al., 1997; Saha et al., 2010; Banerjee et al., 2012; Sappa et al., 2014; Ayers et al., 2016). High arsenic contamination is notably found in regions with hydrothermal mineralization and geothermal activity (Smedley and Kinniburgh, 2002; Webster and Nordstrom, 2003).

The geochemistry of arsenic is influenced by a range of processes, including oxidation-reduction reactions, dissolution-precipitation dynamics, adsorption-desorption mechanisms, and interactions with aquifer mineralogy, organic content, and other environmental factors (Robertson, 1989). Naturally occurring arsenic is released into groundwater due to mineral dissolution from pyrite, garnet, hydrous iron oxides, and sulfide compounds. Additionally, anthropogenic activities such as mining, fossil fuel combustion, arsenic-based pesticides, herbicides, livestock feed additives, and wood preservatives contribute to its mobilization.

Several parameters, including pH, redox potential (Eh), iron concentration, sulfate ions, salinity, temperature, and microbial activity, dictate arsenic speciation and transport within groundwater systems (Bhattacharya et al., 1995; Singh et al., 2015; Rotiroti et al., 2015; Mihajlov et al., 2016; Ahamad et al., 2020). Research has extensively documented arsenic occurrence, mobility, toxicity, and remediation strategies (Bodek et al., 1988; Das et al., 1996; Mandal et al., 1996; Jain and Ali, 2000; Acharyya et al., 2000; Nickson et al., 2000; Matschullat, 2000; Bhattacharya et al., 2002; Bissen and Frimmel, 2003; Mohan and Pittman, 2007; Banerjee et al., 2012; Patel et al., 2016; Ahmad et al., 2017; Olea et al., 2018; Wongsasuluk et al., 2018; Li et al., 2018; Litter et al., 2019; Madhav et al., 2021).

NATURAL SOURCES OF ARSENIC IN INDIA

Researchers suggest a potential link between the geological formations of the Himalayan Mountain range, the Shillong Plateau, and the widespread presence of arsenic in India. Various geological sources within the country contribute to arsenic contamination, including:

- **Eastern India (Rajmahal Basin):** Gondwana coal seams in this region contain arsenic concentrations of approximately 0.02%.
- **Bihar (Mica Belt):** Arsenic levels in the mica-rich belt of Bihar range from 0.08% to 0.12%.
- **Central India (Vindhyan Range):** Shale formations containing pyrite in this range exhibit arsenic concentrations of 0.26%.
- **Son River Valley (Eastern India):** Known for its gold deposits, this valley records an average arsenic concentration of 2.8%.
- **Eastern Himalayas:** Sulfide-rich outcrops in this region contain arsenic levels of around 0.8%.

MAJOR INCIDENCES OF ARSENIC IN GROUNDWATER

Arsenic in groundwater primarily exists in two oxidation states—arsenate (As⁵⁺) and arsenite (As³⁺)—with the ability to transition between these forms through redox reactions. Additionally, arsenic can occur

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in organic forms due to biomethylation, a process facilitated by various organisms, including plants, aquatic life (such as fish and crabs), and even humans. Certain microorganisms, like bacteria and fungi, play a crucial role in converting inorganic arsenic into organic forms through biomethylation.

Arsenic is also present in different oxidation states within sulfide minerals and metal oxides, particularly iron oxides. Studies indicate that regions rich in sulfide minerals, such as Bihar and the Ganga Basin, are major contributors to arsenic contamination in groundwater. High arsenic levels have been detected in wells located in the Indo-Gangetic alluvium to the west and the Brahmaputra alluvium.

During the Holocene period, arsenic became concentrated in sediments through natural precipitation of hydroxides, which absorbed arsenic released from weathered sulfide minerals. Over time, biogeochemical processes and redox reactions facilitated the dissolution of both arsenic and iron oxides into aquifers. Historical records indicate that arsenic contamination in the Ganga-Brahmaputra river basin dates back to the late Quaternary and Holocene periods. Today, various biogeochemical mechanisms continue to drive the mobilization of arsenic in most contaminated water sources.

Arsenic, Geology, and Meteorology of Ballia & Ghazipur Districts

In the geological context, the Ballia and Ghazipur districts feature two distinct types of plains—older and younger alluvial plains—as illustrated in Fig. 1 (Azam, A., 2013). Both plains contain arsenic (As); however, the younger alluvial plain exhibits a higher concentration than the older one. This difference arises from the organic-rich, unoxidized silt and sediment prevalent in the younger plain. Numerous studies have indicated significant toxic contamination in the younger alluvium.

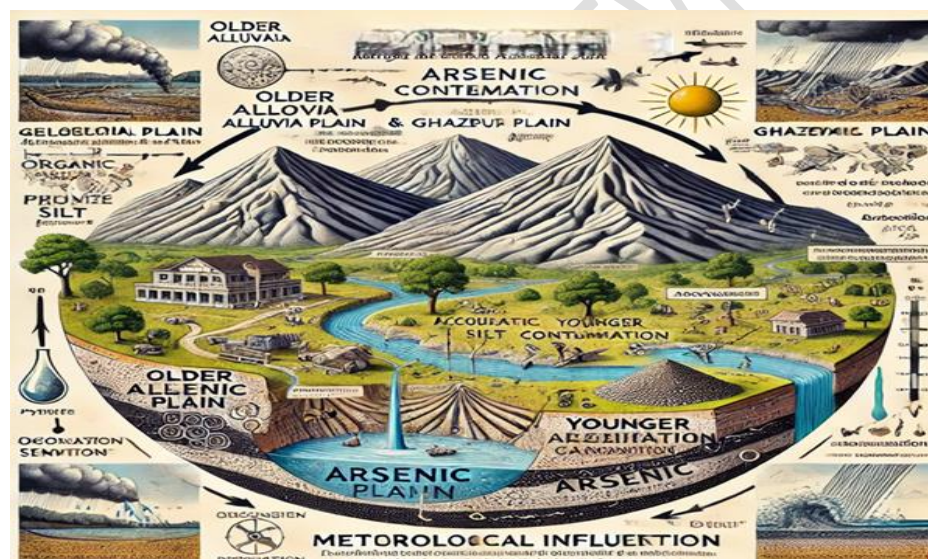


Fig.1. Geological structure of Arsenic mobilization in Groundwater

The primary source of this contamination is linked to pyrite minerals containing arsenic, which are transported via fluvial sediments from the Himalayas to the Ganga River. Over time, these sediments accumulate in the younger alluvial plain, making it a hotspot for groundwater arsenic contamination, particularly in Ballia district.

Seasonal fluctuations in arsenic levels have also been observed between pre- and post-monsoon periods. Following the monsoon season, arsenic concentrations tend to rise due to the influence of rainfall (Izaditame, F., et al., 2022).

MECHANISMS OF ARSENIC MOBILIZATION

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The presence of arsenic-contaminated groundwater in the study areas is not solely attributed to arsenic-rich source rocks. Two major factors contribute to its mobilization:

1. **Biogeochemical Processes** – Arsenic mobilization occurs due to specific geochemical interactions, causing its transition from the solid phase or adsorption sites into groundwater.
2. **Retention in Aquifers** – Arsenic persists in aquifers instead of being flushed out, suggesting that the release rate from source materials surpasses the natural groundwater flushing rate.

Arsenic transport occurs through different hydrogeochemical mechanisms. Under **alkaline and oxidizing conditions**, arsenic undergoes desorption, while **reducing conditions** lead to arsenic dissolution (Fig. 3). The weakening of bonds between arsenic and mineral surfaces further enhances its mobilization (Dzombak, D. A., et al., 1986; Smedley & Kinniburgh, 2002; Anawar, H. M., et al., 2004; Azam, A., et al., 2023).

The conventional understanding of arsenic contamination in groundwater gains a new perspective within the geological context of West Bengal. Researchers (Das et al., 1994) have extensively studied the role of sulphide minerals, particularly pyrite (FeS_2), in the release of arsenic through oxidation. This theory suggests that sulphide minerals, such as arsenopyrite, present in shallow aquifers undergo oxidation, leading to the release of arsenic into groundwater (Mandal et al., 1998).

Another significant factor contributing to arsenic discharge is the depletion of groundwater levels due to excessive irrigation demands. Recent research indicates that arsenic desorption or dissolution from iron oxide may influence its regional distribution in groundwater (Smedley, 2004). This innovative geological approach sheds light on the complex mechanisms responsible for arsenic contamination. A novel geochemical technique now enables the controlled dissolution of iron oxide in reductive environments, thereby increasing arsenic concentrations in water.

Recent scientific studies have also examined the impact of excessive water use in agriculture, particularly in fertilization and irrigation. As a result, phosphate from fertilizers in shallow subsurface water reservoirs has been mobilized, causing the release of arsenic (As) through anion exchange on reactive mineral surfaces. Renowned research institutions, including the National Institutes of Health (NIH) and the Central Ground Water Board (CGWB), along with distinguished scholars such as Majumdar et al. and Michael and Voss, have conducted extensive studies on this issue.

ARSENIC FLUCTUATIONS IN GROUNDWATER: UNVEILING DYNAMIC PATTERNS OVER TIME AND SEASONS

A groundbreaking geological and scientific investigation has revealed significant fluctuations in arsenic (As) concentrations in groundwater across different seasons and time periods. Notably, arsenic levels reached their lowest during the monsoon season, a phenomenon attributed to seasonal variations in groundwater recharge and irrigation depletion effects (MacArthur et al.).

This comprehensive study analyzed arsenic concentration changes in tube wells across various blocks of the arsenic-contaminated Ballia district from 2017 to 2018 (Azam, A. & Kunwar, A., 2018). A clear correlation was established between arsenic content in pre-monsoon and post-monsoon periods. The findings indicated that increased rainfall intensity led to a decrease in arsenic concentration due to enhanced dilution (Farooq et al., 2010). Conversely, during winter and pre-monsoon seasons, arsenic levels increased due to reduced dilution effects.

Groundwater flow patterns were found to be significantly influenced by surface and subsurface redox potential, driven by seasonal rainfall variations. The low-lying shallow basin and frequent flooding in Ballia during the monsoon contributed to rising groundwater levels (Ahamed, S., 2006; Azam, A., 2020). Prolonged waterlogging throughout the year resulted in lower groundwater flushing rates, causing variations in arsenic concentrations between monsoon and pre-monsoon seasons.

Arsenic mobilization in groundwater was controlled by sediment-water interactions and redox conditions. During the monsoon, arsenic was released into groundwater due to the dissolution of Fe(III)-oxyhydroxides, whereas in the pre-monsoon period, arsenic was reversibly adsorbed onto Fe(III)-oxyhydroxides. These distinct geochemical processes played a crucial role in arsenic concentration fluctuations. For this study, multiple sampling sites in Ballia were carefully selected, including Belhari, Bansdeeh, Sikandarpur, Bairiya, Hanumanganj, and Murli Chhapra (India, GU J Sci 2012, 25(4), 853–861p; Katiyar, S. et al., 2014; Azam, A., 2020; Azam, A. & Dr. Sarma, B. K., 2019). **Figures 2 & 3** illustrate these variations in arsenic concentrations across different locations and time frames.

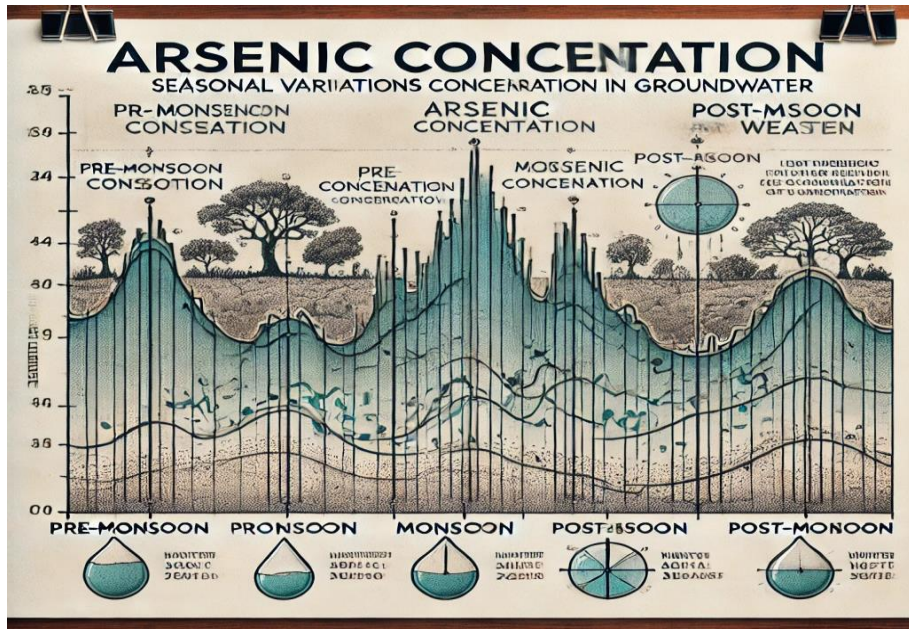


Fig. 2. Seasonal Variation of Arsenic concentration During Pre & Post Monsoon Seasons

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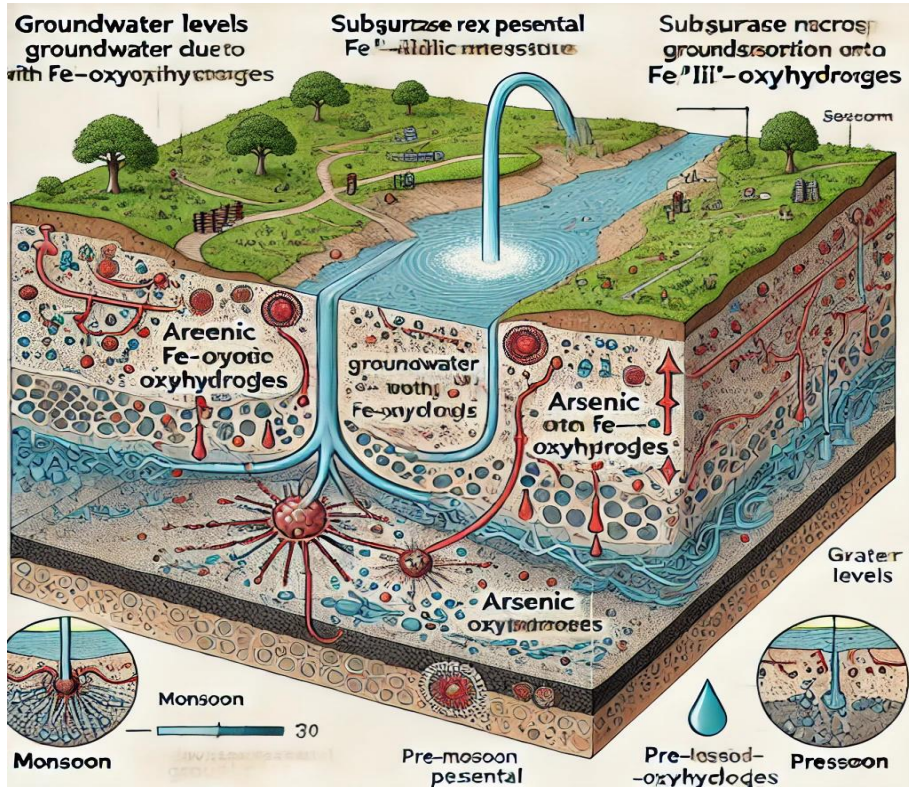


Fig.3. Groundwater enrichment of Arsenic during Pre monsoon season by Fe-OOH redox process

CONCLUSION

Arsenic, a highly toxic element, enters water and soil primarily through river floodplains, where it undergoes mobilization and sedimentation. Even before human intervention, arsenic-contaminated groundwater has been a persistent issue, leading to significant health risks, social disruptions, and economic burdens. The lack of awareness among the inhabitants of the Gangetic Plain regarding arsenic's potential dangers necessitates a thorough investigation to detect its presence in soil and water samples.

This study aims to analyze both organic and inorganic forms of arsenic, establish concentration thresholds across different locations, and examine its speciation. The mobilization of arsenic from rock weathering into water sources and its subsequent sedimentation in affected areas have been extensively studied through mineralogical analyses. While several color-coded indicators have been proposed for arsenic detection, a more precise and reliable method is required.

A geochemical study is fundamental to understanding arsenic's behavior under varying hydrogeological and environmental conditions. Seasonal variations in arsenic concentration are evident, with levels typically decreasing from the pre-monsoon to post-monsoon periods. This fluctuation is primarily influenced by changes in groundwater levels during the rainy season and its extensive use for irrigation. A direct correlation exists between arsenic concentration and seasonal rainfall patterns; increased rainfall enhances dilution, leading to lower arsenic levels, whereas reduced rainfall during the winter and pre-monsoon seasons results in higher concentrations due to decreased dilution. The monsoon

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period thus plays a crucial role in regulating arsenic levels in groundwater, highlighting the significance of seasonal hydrological dynamics in arsenic contamination studies.

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